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Ultrafast Laser Applications in Manufacturing Processes: A State of the Art Review

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ABSTRACT

With the invention of chirped pulse amplification for lasers in the mid-1980s, high power ultrafast lasers entered into the world as a disruptive tool, with potential impact on a broad range of application areas. Since then, ultrafast lasers have revolutionized laser-matter interaction and unleashed their potential applications in manufacturing processes. With unprecedented short pulse duration and high laser intensity, focused optical energy can be delivered to precisely defined material locations on a time scale much faster than thermal diffusion to the surrounding area. This unique characteristic has fundamentally changed the way laser interacts with matter and enabled numerous manufacturing innovations over the past few decades. In this paper, an overview of ultrafast laser technology with an emphasis on femtosecond laser is provided first, including its development, type, working principle, and characteristics. Then ultrafast laser applications in manufacturing processes are reviewed, with a focus on micro/nano machining, surface structuring, thin film scribing, machining in bulk of materials, additive manufacturing, bio manufacturing, super high resolution machining, and numerical simulation. Both fundamental studies and process development are covered in this review. Insights gained on ultrafast laser interaction with matter through both theoretical and numerical research are summarized. Manufacturing process innovations targeting various application areas are described. Industrial applications of ultrafast laser based manufacturing processes are illustrated. Finally, future research directions in ultrafast laser based manufacturing processes are discussed.

1. INTRODUCTION

Ultrafast lasers, or more precisely known as ultrashort pulse lasers, utilize mode locking to produce pulses with femtosecond (fs) or picosecond (ps) durations [1]. Because this paper concerns ultrafast laser applications in manufacturing processes that involve strong laser-matter interaction with typical electron-phonon coupling on a timescale of picoseconds [2], femtosecond lasers will be emphasized in this review. A fs laser when used in manufacturing modifies or ablates materials with high intensity ultrashort pulses of < 1 ps duration to create high precision features at the micro and nano scale in a highly localized and controlled manner. Although nanosecond (ns) and

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ps lasers have been widely used in the field of laser micromachining, fs lasers offer superior quality in micromachining and are often indispensable in nano manufacturing. With rapid development in fs laser technology, especially the maturity of economical and reliable fs fiber lasers, more fs lasers will be used in the future. Hence, this paper aims to review the progress made in the past mainly in fs laser based manufacturing processes and point out emerging application areas and directions for potential future research. Because of the vast literature devoted in this field in the past few decades, this review attempts to provide an insightful overview of representative work in each topic area without losing the broadness in scope. In addition, due to the huge variety of manufacturing processes involving fs lasers, this review will mainly focus on fs laser based subtractive and additive processes.

2. A BRIEF HISTORY OF ULTRAFAST LASER DEVELOPMENT

The pursuit of ultrashort pulse durations traces back to the 1960s, soon after the first demonstration of a ruby laser by Theodore Maiman [3]. Pioneering mode locking research of solid state and organic dye lasers was reported in the 1960s and 1970s, including both experimental investigations [4, 5] and theoretical studies [6]. A turning point in ultrafast laser development came with the report of a sub 100fs self-mode-locked Ti:sapphire laser by Spence et al. in 1991 [7], while demonstrating the advantage of solid state lasers over organic dye lasers in practicality and efficiency [8]. Subsequent rapid advances in the 1990s witnessed a continuous drop of pulse duration to as short as below 10 fs [9]. While ultrashort pulses directly from a laser oscillator with nanojoule energy at megahertz repetition rates may be more applicable for information processing and communications, highly energetic pulses are required for heavy duty micro/nano materials processing in manufacturing.

In 1985, a revolutionary development was made by Strickland and Mourou with the invention of chirped pulse amplification (CPA) to create ultrashort, ultrahigh intensity laser pulses [10], for which they were awarded a Nobel Prize in Physics in 2018. The CPA approach first stretches the mode-locked low power ultrashort pulses in time by a factor of ~10^4 from its original duration using gratings. Then these long pulses with much lower peak power are safely amplified by several orders of magnitude while avoiding damage to the gain medium through nonlinear processes. Finally, these amplified pulses are recompressed in time to their original pulse duration, achieving orders of magnitude higher peak power than an ultrafast laser can generate. Without question, CPA removed a tremendous roadblock in the quest for high power ultrafast lasers by enabling the efficient amplification of ultrafast pulses to high energy within a compact package and thus opened a new era in ultrafast laser development. Now CPA lasers are widely used in the world for both scientific research and industrial applications, with average power exceeding 100 W [11], peak power reaching terawatt levels [12], pulse duration dropping below 10 fs [13], repetition rate reaching megahertz [11], and wavelength covering extreme ultraviolet to mid-infrared [14, 15].

As another class and an important addition to the portfolio of ultrafast laser source, ultrafast fiber lasers started to attract attention in the 1980s with the development of fiber lasers and fiber amplifiers [16]. Early interest in ultrafast fiber lasers was driven by the demand for shorter pulses in communications. For industrial applications such as materials processing, high average power fs fiber lasers were developed utilizing CPA and large-mode-area hollow core photonic fiber technology [17]. Femtosecond fiber lasers generally use Yb-, Er- or Tm-doped gain media and emit in the near infrared regime. A range of commercial femtosecond fiber lasers are now available which can operate at <100 fs pulse duration, megahertz repetition rate, gigawatts peak power, and kilowatt level average power. The potentially high throughput and low cost of ownership make femtosecond fiber lasers attractive in industrial applications.

3. ULTRAFAST LASER MACHINING CHARACTERISTICS

3.1 Ultrafast laser-matter interaction

Ultrafast lasers possess three key distinctive properties compared to lasers with longer pulse durations: ultrashort pulse duration, ultrahigh intensity, and large spectral bandwidth. The first two properties in particular give rise to some unique physical phenomena enabled via ultrafast laser-matter interaction in manufacturing applications. Because of their <1 ps pulse duration, ultrashort pulses in principle do not allow heat to diffuse into the surrounding area and thus prevent large thermal damage. Figure 1 illustrates somewhat in an ideal way the drastic differences between long pulse and ultrashort pulse laser micromachining [18]. For long pulses (e.g., >1 ns), the material has time to be heated up and melt at the focal volume and to diffuse into the surrounding material. A portion of the molten material is vaporized and ejected as high-velocity microdroplets while the rest of the melt re-solidifies, resulting in poor machining quality that is characterized by a heat-affected zone (HAZ), recast, dross, and microcracks. In contrast, for ultrashort pulses (e.g., <1 ps), the thermal diffusion length is generally much smaller than the laser penetration length. Hence, ultrashort pulses induce rapid ionization and directly turn the material at the irradiated volume into a mixture of plasma, vapor, and nanodroplets with subsequent ejection. Negligible melting and heat diffusion into the surrounding lead to a highly clean and precise machining quality that is free of the defects associated with longer pulse durations.

Some new phenomena in ultrashort pulse-matter interaction with dielectrics stem from the ultrahigh intensity that a focused pulse can reach (>10^{18} W/cm^2, the relativistic regime). As laser intensity is above a threshold value, a normally transparent material begins to absorb laser radiation through multiphoton absorption, which is responsible for generating some initial seed electrons from a dielectric or semiconductor material for the subsequent avalanche ionization, plasma formation, and material breakdown. Since multiphoton absorption is a nonlinear process that does not rely on seed electrons provided by defects or contaminants, the breakdown threshold for a material is definite and can be accurately determined. This deterministic nature of ultrafast laser-matter interaction lends itself to numerous new applications in manufacturing processes. For example,

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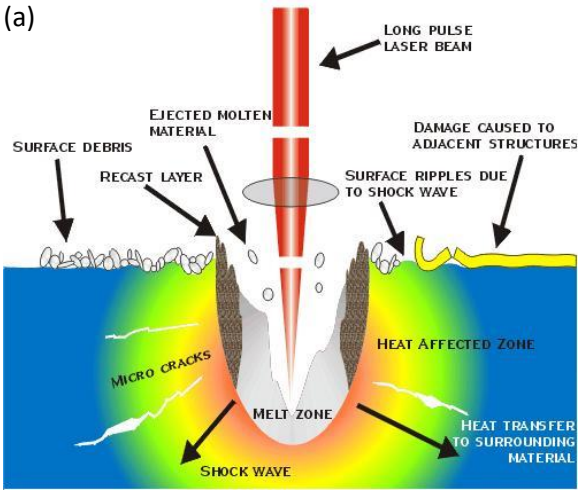
nanoscale features are made possible without a nanoscale focal spot size. This diffraction limit defying performance can be achieved by controlling the Gaussian intensity profile in such a way that only a small portion of the beam in the middle of the spot reaches the threshold intensity, effectively reducing the useful portion of the beam to below the wavelength value and thus submicron/nanoscale features can be created on the target sample. Another fascinating phenomenon owing to this nonlinear laser-matter interaction is the ability to access inside a material. This is usually done by tightly focusing a laser beam into a material with a high numerical aperture objective lens, so that only the focal volume inside the material reaches the damage threshold.



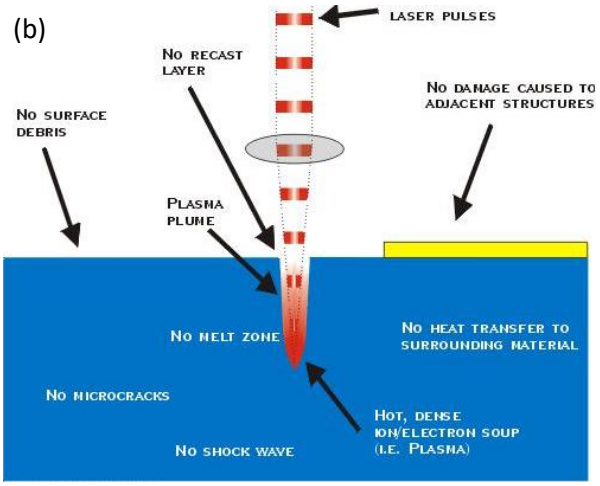


Fig. 1 Illustration of laser-matter interaction [18]: (a) long pulse and (b) ultrashort pulse.

3.2 Typical experimental setup for ultrafast laser machining

Figure 2 shows a typical ultrafast laser machining setup, which consists of the following main modules: ultrafast fs laser system, beam shaping, beam control, beam delivery and focusing, process monitoring, motorized 3D positioning stage, and computer control. Most of the current ultrafast fs lasers use Ti:sapphire as a gain medium because of its broad bandwidth. Low energy (~nJ) and high repetition rate (~100 MHz) ultrashort pulses are first generated in a mode-locked oscillator pumped by a solid state laser. These pulses are then amplified through the technique known as chirped-pulse amplification to microjoule and millijoule level for machining applications. Typical commercially available systems can produce pulses with tens of fs duration at about 800 nm wavelength and up to 10 kHz repetition rate. The average output powers can reach several watts. This power output does not allow a large amount of material removal and hence fs laser is most suitable for micro and nano machining applications.

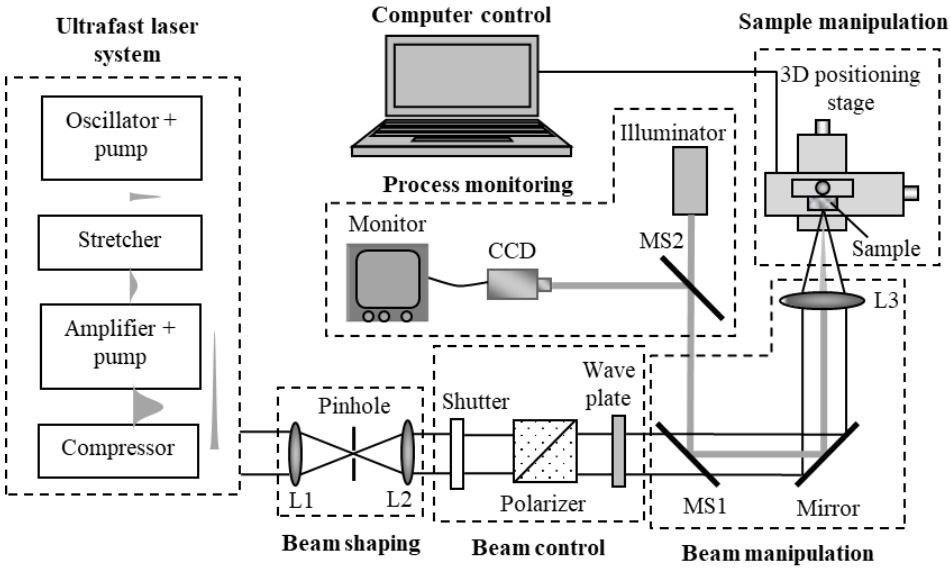


Fig. 2 A typical setup of femtosecond laser machining system showing various functional modules.

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If the ultrashort pulses from the fs laser have fluctuations superimposed on a Gaussian intensity profile, a spatial filter as shown in Fig. 2 can be used to improve the beam quality by suppressing the noise and smoothing the beam profile. Also, wavefront correction techniques have been used to achieve high peak intensities at the focal spot [19]. In the beam control module, a high speed shutter is used to control the number of pulses. Pulse energy is adjusted with the combination of a polarizer and a half waveplate. Additional optics can be used to control the polarization state of the laser beam. In micromachining, fast laser scanning is often needed, which can be achieved in the beam manipulation module. Commercial laser beam scanning heads are available for this purpose. Figure 2 only illustrates simple beam delivery and focusing optics. The control of sample movement is done by the 3D positioning stage. Quality control is realized through the in-process monitoring module. One approach is through the use of a separate illumination source and CCD camera as shown in the figure. Another common practice is to use the reflected light from the ablation area to observe the processing site on the sample. Finally, the fs machining system is controlled by a computer with necessary application software for controlling various devices. There are several parameters to be controlled for fs laser machining: pulse energy, pulse duration, repetition rate, focal spot size, and polarization. In addition, sample movement and beam scanning need to be controlled based on the machining requirements.

4. APPLICATIONS OF ULTRAFAST LASER IN MANUFACTURING PROCESSES

4.1 Micro/nano machining

Ultrafast lasers are promising tools for ultrahigh precision machining due to the ultrafast laser absorption, heat transfer, material removal, and thus minimized heat-affected zone (HAZ) formation. It is usually considered as a "cold" ablation process. It was demonstrated that femtosecond laser machining is much cleaner and smoother with sharp edges compared with machining by nanosecond laser pulses (Fig. 3 [20]). The ultrahigh laser intensity also enables it to process a rich diversity of materials, including brittle transparent materials by nonlinear absorption.

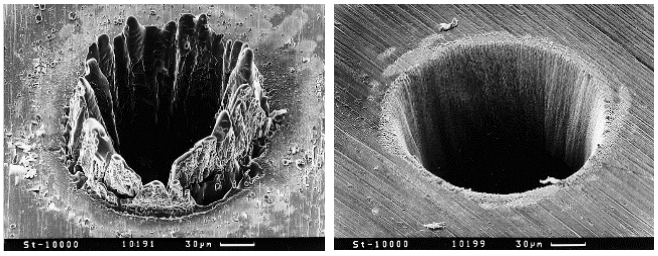

Fig. 3 Drilling by nanosecond laser (left) and femtosecond laser (right) [20]. The scale bar is 30 μm.

4.1.1 Micromachining

(a) Laser ablation

Laser micro/nanomachining is accomplished by laser ablation, which refers to the material removal process resulting from laser irradiation. Despite the property difference between different materials, it was shown that different materials behave similarly under the irradiation by femtosecond pulses at fluences below the threshold for plasma formation [21], implying some very general mechanisms are responsible for the material removal process. It has been widely accepted that the possible ablation mechanisms include spallation [22, 23], phase explosion [22-24], critical point phase separation [25], and fragmentation [22, 26]. Spallation is caused by laser-induced tensile stress, which can cause the formation of voids and defects when its amplitude exceeds the matter bonding, and trigger the consequent decomposition. Phase explosion, also known as explosive boiling, refers to the explosive decomposition of metastable material into a mixture of vapor and liquid when the material is superheated to temperatures close to the critical temperature. For critical point phase separation, the material is also rapidly heated to a temperature above critical point. During the fast expansion, the material is pushed into the metastable region by crossing the critical point, leading to a phase separation into an ablated vapor phase and an unablated solid/liquid phase. Fragmentation is the removal of small fragments due to laser-induced thermo-elastic stress, and resultant nonuniform strain rates inside the material. The occurrence of these mechanisms depends on the incident laser parameters and target material properties. Generally, from low to high laser fluence, the dominating mechanism switches from spallation, to critical point phase separation, and to fragmentation. In addition, there is non-thermal ablation, such as Coulomb explosion induced by strong electric field near the surface [27-30]. It should be pointed out that although the ablation mechanisms discussed above occur and dominate in different fluence ranges, in many cases, two or more will take place simultaneously. In addition, processing environment also affects laser ablation mechanisms. For example, Kudryashov et al. recently reported the damping of vapor/plume expansion in fs laser ablation of gold surface in isopropyl alcohol due to liquid confinement [31]. Cao et al. demonstrated that enhanced chemical reaction and electronic conductivity were responsible for the change of surface morphology when comparing fs laser ablation of silica in air and etching solutions [32].

(b) Effect of processing parameters

Ablation depth (per pulse) is defined as the depth of the crater ablated by one laser pulse, which is an important quantity to measure the efficiency of laser machining. It can be affected by various laser parameters, target material properties, and ambient environment.

Pulse duration: It was found that with the same laser fluence, the ablation depth decreases with pulse duration [33, 34]. However, when the pulse duration is lower than 1 ps, the ablation depth does not change any more. This is because in this range, the pulse duration is lower than the electron-phonon relaxation time, and the thermal exchange is negligible during the laser irradiation [33, 35-37].

Laser fluence: Laser fluence is another key factor to determine the ablation depth. It was observed in [35] that two ablation regimes exist during ultrafast laser ablation of copper, and the ablation depth (L) depends logarithmically on laser fluence in each regime:

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$$L = \delta ln(\frac{I}{I_{th}^{M}}) \text{ (low fluence)}, \quad (1)$$

$$L = lln(\frac{I}{I_{th}^{H}}) \text{ (high fluence)}, \quad (2)$$

where δ is the optical penetration depth, l is the electronic heat penetration depth, I is the laser fluence, I_{th}^{M} and I_{th}^{H} are the ablation thresholds for each regime. The existence of the two regimes is mainly due to the switch of the dominating energy transfer mechanism from optical penetration to heat diffusion. The phenomena was found to be universal for many other materials [38, 39]. A third regime at even higher fluence was reported for some materials due to the collisionless absorption in vacuum and the early plasma absorption in air [36, 40].

Laser-induced plasma: During laser ablation, the ablated materials can be partially or fully ionized, forming laser-induced plasma. Different from longer pulse ablations, this plasma (generated after hundreds of ps [41, 42]) does not have any interaction with the incident pulse due to the ultrashort pulse duration. Therefore, the plasma shield effect is usually ignored for ultrafast laser machining. However, due to its extremely high laser intensity, an ultrafast laser pulse can create plasma in the ambient fluid by both direct photoionization and ionization by ejected electrons from the target surface. This plasma is called "early plasma" since it can be formed within tens of femtoseconds [43-45], and further absorb the incident laser energy. At high laser fluence, the early plasma can absorb up to 20% of the incident laser energy and thus reduce the ablation depth [43, 44].

Multi-pulse ablation: Laser irradiation by multiple pulses is needed for many laser machining tasks. It has been shown that compared with the single-pulse ablation, the threshold fluence by multi-pulse ablation can be reduced, and the average ablation depth per unit incident fluence will be increased [46, 47]. This phenomena in generally is called the incubation effect, and could be caused by thermal accumulation [48], and enhanced absorption by pre-pulse surface modification [49], formation of micro-and nano-structures [47], and laser induced defect formation [50].

When the time interval between pulses is reduced to extremely short (fs-ns), new mechanisms will come into play, and completely different ablation behavior will happen. It has been revealed that the ablation depth of metals will be suppressed by double-pulse ablation with ultrashort pulse intervals [51, 52], possibly due to the formation of a second shock wave [53], the drop of electronic heat conductivity at high temperature [54, 55], and the plasma shielding effect [52]. Different from metals, ablation of silicon will be enhanced by the same technique, due to electron excitation by the first pulse to enhance the absorption of the second one [33,35–37][54, 56-58].

(c) Processing techniques

Several processing techniques have been proposed to improve the micromachining efficiency and quality. Fig. 4 shows the typical drilling strategies [59]. Single pulse drilling is fast, but the achievable drilling depth is very limited. Percussion drilling uses laser pulses which move sequentially along the drilling direction, and deeper holes can be drilled. Trepanning combines percussion drilling with the circular movement of the laser beam relative to the target. Helical drilling is very similar to trepanning drilling, and the only difference is that the laser beam moves helically. The advantage of helical drilling over trepanning is that the machining position can always be maintained at the laser focal spot, and thus helical drilling suits better for deeper holes. These strategies can be easily modified to be suitable for other machining processes like scribing and cutting.

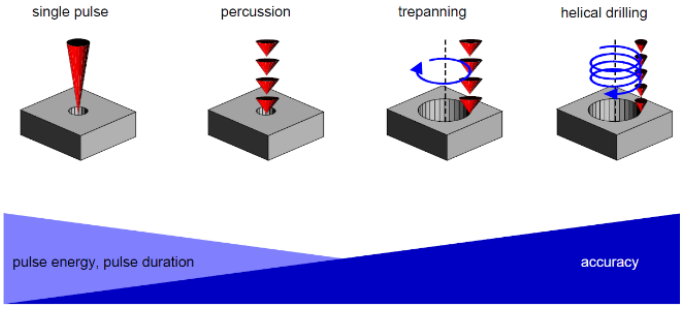

Fig. 4 Process strategies with increasing accuracy and decreasing pulse energy/duration, from left to right [59].

(d) Micromachining of various materials

Ultrafast lasers have been used for micromachining of various materials, such as metals [60-63], semiconductors [64-67], glass [68], organic materials [69], and composite materials [70], as shown in Figs. 5 and 6. It has the capability for deep-hole drilling, cutting, surface scribing, microstructuring, trimming, etc. Regardless of the material, machining quality and precision are very high with sharp edges and no cracks or debris. Ultrafast laser micromachining can be used in various applications, such as fuel injector nozzle drilling, stent fabrication, micro-device manufacturing, etc. In reference [71], it demonstrates that femtosecond laser possesses advantages for fuel injector nozzle drilling in edge sharpness, HAZ, and resolution compared with water-jet guided μs-laser and micro-EDM.

The effects of processing conditions on machining quality and efficiency have been extensively studied for metals and semiconductors. It was found that by using shorter duration pulses, laser-induced thermal effects and HAZ will be suppressed. When the pulse duration is shorter than the electron-phonon relaxation time, clean ablation with minimized HAZ will be achieved [72]. However, with very short pulses, the laser fluence should be kept relatively low to avoid nonlinear beam profile distortion [73, 74]. This issue can also be overcome by adopting vacuum environment leading to the absence of laser-air interaction [60-62]. With a too high laser fluence, although machining efficiency is high due to the high ablation depth, HAZ will also be increased which lowers the machining quality, losing the benefit of ultrafast laser machining. Therefore, a low laser fluence (slightly higher than the threshold) with a high repetition rate is a preferred combination for many cases [75, 76]. Laser polarization was revealed to be important to control the drilling quality. Circular polarization is needed to drill a through-hole with a round exit [64, 65].

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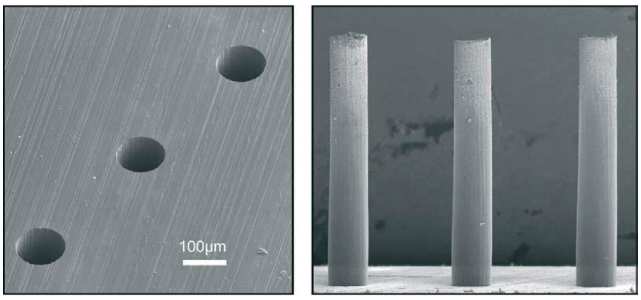

Fig. 5 Micro-holes drilled in a 1mm thick stainless steel plate [62]

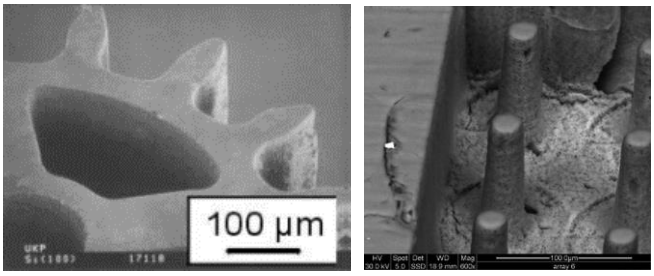

Fig. 6 Micromachining of silicon [67] (left) and bovine cortical bone [69] (right).

Ultrafast lasers were also found capable of fabricating high aspect ratio and high quality microchannels in transparent materials which is difficult, if not impossible, using traditional machining techniques, due to the brittle structure and low thermal conductivity of the material. The first developed method was the laser-assisted chemical etching. The target was irradiated by femtosecond laser pulses, followed by heat treatment and dilute aqueous hydrofluoric solution etching [77, 78]. The same technique without heat treatment was used to create microchannels with a high aspect ratio (50:1) [79-82]. The disadvantage of this technique is the requirement of a time consuming post-processing by toxic chemicals. An alternative microchannel fabrication method is direct laser drilling, which is fast with no post-processing [83-85]. It was found that drilling from the rear surface generates deeper holes than in the front surface, as the plasma formed by drilling from the rear surface is much weaker [85]. Distilled water and other liquids were reported to be helpful to raise the aspect ratio by reducing the redeposition of ablated materials [84, 86]. Ultrasonic agitation [84] and post-annealing [87] were also found effective in improving the drilling process in terms of aspect ratio. A comparison of the air and vacuum drilling processes found that vacuum drilling increased the aspect ratio to over 100:1 in [88].

4.1.2 Nanomachining

Ultrafast laser ablation shows great advantages in minimizing the HAZ formation, and has demonstrated superior capability in high precision micromachining. In principle, it possesses the potential for even higher resolution machining – nanomachining. In fact, the barrier for its application in nanomachining is not the HAZ, but the diffraction limit of the laser spot size. In general, the focused laser spot size is limited by its wavelength, which cannot satisfy the requirement of nanomachining. To overcome this limitation, several methods have been successfully developed.

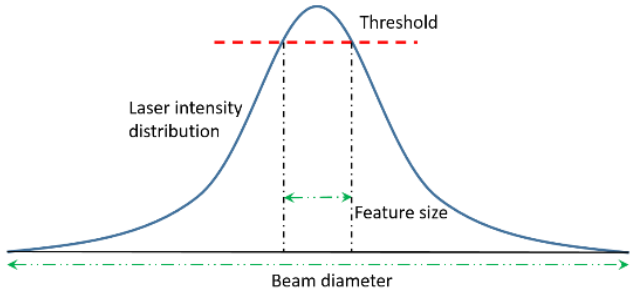

Fig. 7 Schematic of nanoablation by Gaussian beams.

One method to achieve the ultrahigh fabrication resolution beyond the diffraction limit is to accurately control the peak laser intensity slightly higher than the processing threshold. Ultrafast laser ablation has deterministic thresholds for different materials, meaning that the material can be processed only when the laser intensity is higher than a certain threshold value. Therefore, by using a Gaussian beam, higher fabrication resolution can be obtained by a low laser intensity where only the central part of the laser beam exceeds the threshold to induce ablation (Fig. 7). By using this method with femtosecond lasers from infrared (IR) [89], visible [90], to ultraviolet (UV) [91] and extreme UV [92], sub diffraction limit nano-features were successfully fabricated in different materials. With this method, nanomachining can be achieved with simple setup and simple objective lenses. This method was improved by using a double-pulse setup (one IR pulse and one UV pulse with controlled interval), which can reduce the ablation threshold of fused silica by 90% [93, 94] (Fig. 8). In [95], by adopting temporal airy pulses, high aspect ratio (30:1) nanochannels with diameters of hundreds of nanometers were fabricated. In principle, taking advantage of the threshold effect has no limit in the machining resolution, and features as small as 15 nm have been fabricated [90]. However, to achieve higher resolution, higher laser intensity control accuracy is required, which demands extremely stable laser energy output from the laser machines. In addition, the pulse-to-pulse energy fluctuation significantly limits the repeatability of this process with high resolution. Therefore, the reproducible fabrication resolution is usually limited to over 100 nm [92].

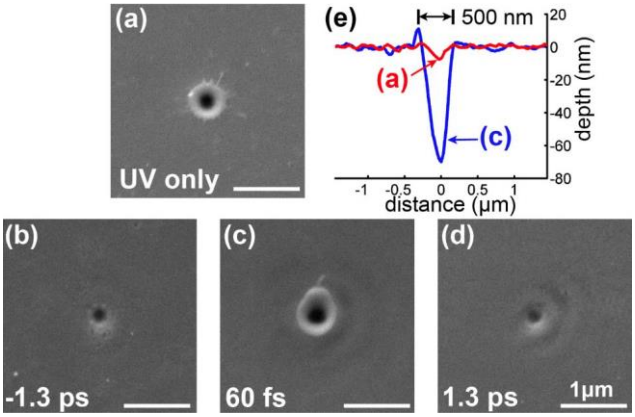

Fig. 8 Nanoholes ablated by (a) UV, and (b-d) UV-IR pulse train with different delays [94].

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To achieve ultrahigh resolution far beyond the diffraction limit, another direction is to overcome the diffraction limit by novel beam focusing techniques. By applying femtosecond laser beams in optical near fields, such as scanning probe microscopy [96], near-field scanning optical microscopy [97], plasmonic nanolithography [98-100], microlens array nanolithography [101, 102], and interference lithography [98, 103], nano-features of tens of nanometers were successfully fabricated on various material surfaces including metals, semiconductors, and dielectrics.

As discussed in Section 4.1, ultrafast laser irradiation can create self-organized periodic nanostructures on surfaces. It is attributed to the periodic laser energy spatial modulation during laser irradiation. It was observed that by carefully controlling the laser intensity to be slightly higher than the ablation threshold, periodic nano-grooves can be created on a surface by single pulse irradiation [104-107]. Furthermore, by adjusting the laser intensity, it is possible to control the number of nano-grooves, and only one nano-groove with a width of tens of nanometers can be fabricated with low laser intensity [106], as shown in Fig. 9.

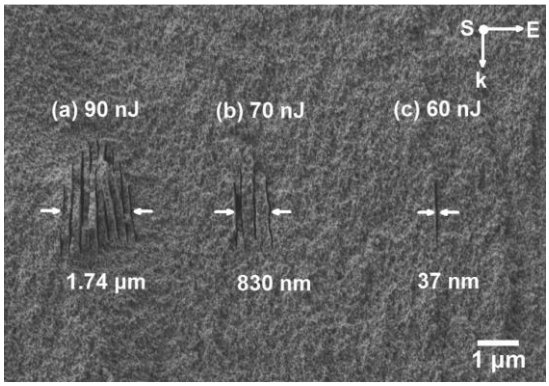

Fig. 9 Evolution from nanovoid array to single nanovoid with decreasing laser intensity [106].

4.2 Surface structuring

4.2.1 Micro/nano surface structures produced by fs lasers

Femtosecond laser has the ability to create either ordered or random surface structures at both micro and nano scale. The most commonly observed structure is laser induced periodic surface structure (LIPSS), also known as surface ripples. Although femtosecond lasers are routinely used to create LIPSS these days, this phenomenon was discovered more than 50 years ago by Birnbaum [108] with a ruby laser. Subsequent studies in the early days found similar periodic structures using various types of lasers [109-111]. With the emergence of high power fs lasers in the 1980s, surface structuring has shifted to the use of fs lasers and attracted worldwide attention. Today it remains an active research area with numerous new developments being reported over the past ten years, which will be the main focus of this part of the review. Among the many different structures produced using fs lasers, LIPSS are clearly the most widely studied one. LIPSS seems to be a universal phenomenon for a variety of materials including metals [112-115], semiconductors [116-118], polymers [119], and dielectrics [120]. There are two types of LIPSS being reported so far: low spatial frequency LIPSS (LSFL) and high spatial frequency LIPSS (HSFL). Figure 10 shows some examples of LIPSS structures found for different materials. The LIPSS structures are generally oriented perpendicular to the laser beam polarization direction for metals and semiconductors, and for dielectrics the orientation can be both perpendicular and parallel [121, 122]. By changing the laser beam to elliptical polarization, the ripples can be rotated; for circularly polarized beam, uniformly distributed bumps can be produced. The dependence of LIPSS orientation can also be changed with a double beam irradiation scheme. With precisely controlled pulse delay of <1 fs resolution, i.e., by controlling the phase difference of the two incident pulses, the LIPSS formed on silicon was rotated by 90° [123]. In another study using double beam irradiation, it was found that the polarization of the first pulse decides the LIPSS orientation for fused silica while for silicon the stronger pulse controls the orientation [120].

Under single beam irradiation conditions, the spatial period of LSFL is close to the laser wavelength, but it can be smaller or larger depending on laser wavelength, incident angle, laser fluence, and material type [113, 120, 124]. Ambient air pressure was also found to affect the LSFL period in a study by Nivas et al., in which the ripple period on silicon decreases with increasing air pressure [125]. Using a double pulse irradiation of nearly equal energy, Höhm et al. showed that time delay can reduce the LSFL period on silicon from 790 to 550 nm while the period remains nearly constant at about 760 nm for the same delay range [120]. Another study with two color fs pulses (800/400nm) showed that for titanium the LSFL orientation and spatial period are determined by the second pulse [126]. Unlike LSFLs that occur for all major material types, HSFLs appear to be a phenomenon for dielectrics and semiconductors only. HSFLs have spatial periods much smaller than incidence laser wavelengths and are usually produced using ultrafast lasers below a material's ablation threshold [127]. They are often very sensitive to the number of laser shots and laser fluence [121, 128]. A recent study by Beltaos et al. showed that, with proper irradiation condition, sub-100 nm HSFL on multi-layer graphene can be produced using a typical fs laser of 840 nm wavelength [129].

Various theoretical and numerical studies have been reported to explain the experimental observations of LIPSS formation, especially for LSFLs [108, 109, 124, 130-134]. The most widely accepted one was developed by Sipe et al. [130] and it shows that inhomogeneous energy absorption as a result of the interference between the incident beam and the surface scattered electromagnetic wave field due to surface roughness is responsible for the formation of LIPSS structures. Later, Bonse et al. combined the Sipe model and Drude model in their study and attributed the formation of LSFL to the interference of the defect-initiated surface plasma polaritons with the incident laser beam [124]. A further advancement was made by He et al. by extending the Sipe model to include excited states for materials like silicon [133]. Their simulation successfully revealed in real spatial coordinate ripple bending and bifurcation, two rarely investigated characteristics of LIPSS. A notable contribution was made recently by Gnilitskyi et al. [134] with the creation of long, highly uniform LIPSS (Fig. 10e) and the discovery of its origin. They found that the LIPSS regularity is governed by the decay length of the excited surface

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electromagnetic waves, and the shorter the decay length the more uniform the LIPSS is. Despite the successful theoretical development for LSFLs, the existing theories for LSFLs can not explain the formation of HSFLs [122]. Based on some studies on HSFL using 800nm fs lasers, it seems that the second harmonic at 400nm plays an essential role in forming HSFLs [121, 128, 135]. More work is definitely needed in this area.

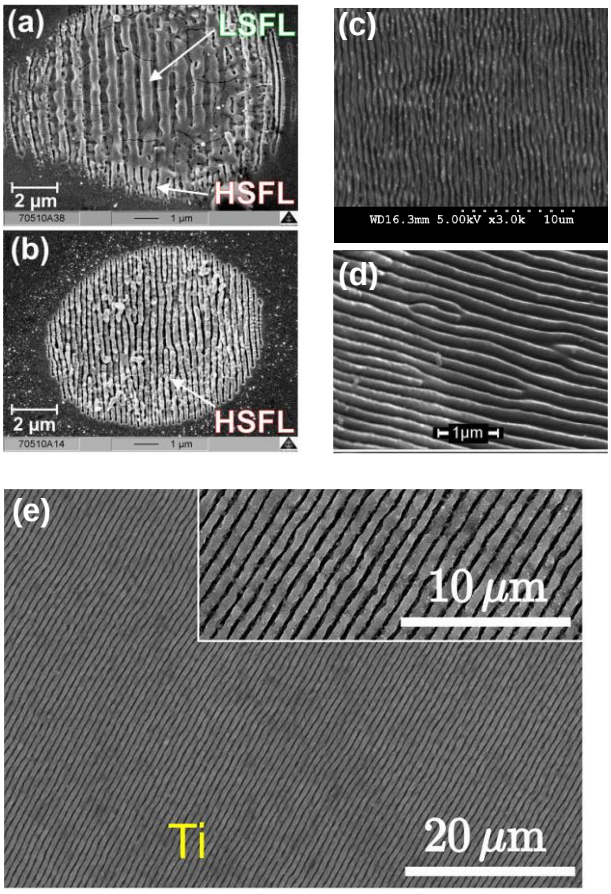

Fig. 10 LIPSS produced using fs laser: (a, b) LSFL and HSFL on ZnO [121], (c) LSFL on silicon, (d) HSFL on diamond [127], and (e) highly regular LSFL on titanium (adapted from [134] on the basis of open access).

Another widely known fs laser induced ordered surface structure is micro cones/spikes. Figure 11 shows the spikes formed on silicon in the environment of SF_6 (Fig. 11a) and in water (Fig. 11b) and the conical structures on titanium (Fig. 11c) and steel (Fig. 11d). According to Carey et al., the spikes on silicon are formed after tens or hundreds of laser shots at about 100 fs pulse duration and medium fluence levels (e.g., 1-3 J/cm^2) [136]. The role of the first few laser pulses is to induce melting, vaporization, and/or chemical reaction at some selected sites based on the initial surface morphology, resulting in micro craters/ripples scattered over the surface. When the subsequent laser pulses strike this roughened surface, the incident laser light bounces off the sides of the protrusions and cause preferentially more absorption in the valleys than on the tips of the protrusions. Therefore, more ablation takes place in the valleys and sharp conical microstructures form after certain number of shots. Generally speaking, the necessary conditions for micro spike/cone formation involves relatively high laser fluence and a large number of laser shots (e.g., hundreds), and normally LIPSS is created first after a small number of shots before the transition to conical structures [137, 138]. Interestingly, as micro cones grow with laser irradiation, LIPSS and random nanostructures can be found on the conical surface [138, 139].

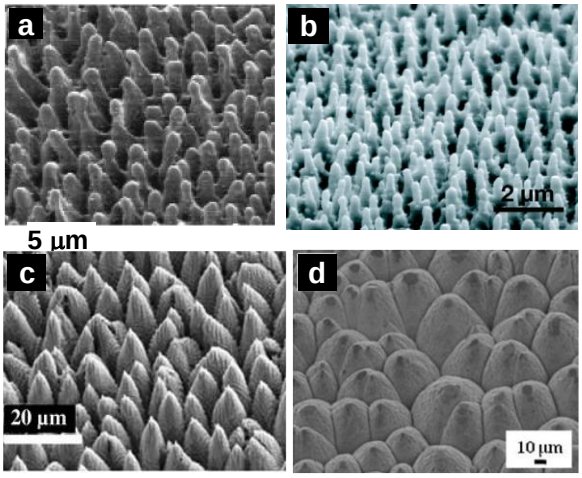

Fig. 11 Laser induced micro cones/spikes on (a) silicon in SF_6 [136], (b) silicon in water [140], (c) titanium in air [139], and (d) stainless steel in air [138]

Besides the above-mentioned regular and periodic microstructures, many different types of surface patterns and structures have been demonstrated through beam shaping and multiple beam strategies, as shown in Fig. 12 for some interesting examples. Using a two color (800/400 nm) fs pulses with controlled delay, dot-like matrix structure is created on Mo plate (Fig. 12a) [141]. The new pattern is not formed by a simple overlap of two sets of LIPSSs by the two individual beams, but rather through interaction of surface dynamics induced by the two beams. Figure 12b shows a feature created on Ni film with a donut shaped, cylindrical vector fs laser beam that is azimuthally polarized [142]. A similar surface pattern is produced on silicon by Nivas et al. [143] using an optical vortex beam. Skoulas et al. demonstrated that a controlled scan of this type of beam enables creation of complex biomimetic structures exhibiting a combination of micro and nano scale features [142]. Periodic arrays of nanotriangles are generated on tungsten using two cross-polarized collinear fs laser beams (Fig. 12c) [144]. The formation of nanotriangles is attributed to the interaction of three sets of periodic nanogrooves triggered by the excitation of surface plasmons from the two time-delayed fs pulses. With a single shot of four interfering fs laser beams at each spot, bump arrays on a gold film are formed as shown in Fig. 12d [145]. The interesting thing is that with increasing laser fluence, a bead appears first on the bump, then followed by a hole to finish. Recently Oliveira et al. reported fs laser surface patterning of carbon fiber-reinforced polymer using laser interferometry [146]. Selective removal of the epoxy resin in the composite material was achieved with controllable pattern period and amplitude.

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Other beam shaping techniques for surface structuring include the use of blazed gratings and spatial light modulator (SLM). High quality and more uniform LSFLs on a few metallic materials are produced using spatiotemporally modified fs pulses with a grating pair [147]. Almeida et al. [148] used a liquid crystal SLM at the Fourier plane of a folded 4f zero-dispersion compressor to transform a 40 fs pulse to a train of several fs sub-pulses with an adjustable sub-pulse separation. It was found that LIPSSs produced by this pulse train are more pronounced with the largest enhancement in ripple amplitude obtained at a sub-pulse separation of 128 fs. Computationally efficient and accurate holograms are generated to produce diffractive multiple beams for parallel processing using SLM. Aided with a galvo scanning system, large area microhole patterns and 3D chessboard like structures are created on Ti6Al4V [149].

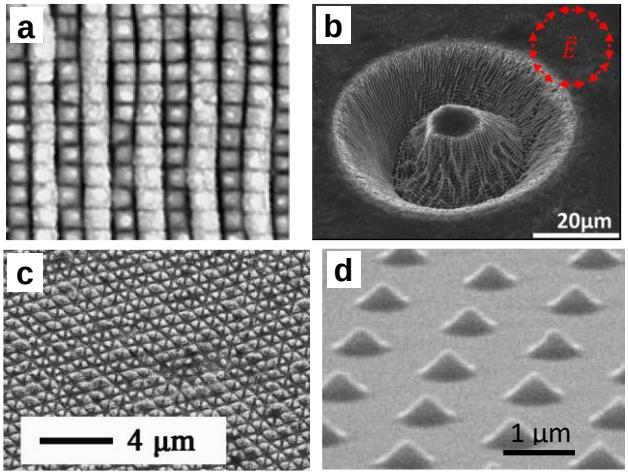
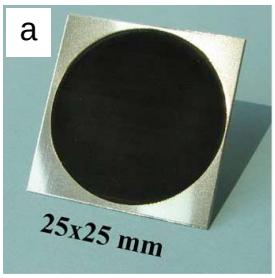


Fig. 12 Various surface structures induced by (a) two-color fs pulses (adapted from [141] on the basis of open access), (b) fs pulse of azimuthal polarization (adapted from [142] on the basis of open access), (c) double fs pulses (adapted from [144] on the basis of open access), and (d) four interfering fs pulses [145].

4.2.2 Surface property and potential applications

Femtosecond laser surface structuring not only alters a material's surface morphology but often imparts some new functionality and properties to the surface, notably optical, mechanical, and chemical properties. Femtosecond laser textured silicon covered with micro spikes, often known as black silicon, is among the early findings to demonstrate nearly 100% absorptance in the visible range, and the spectrum can be extended to 2.5 μm when processed in an ambient SF6 gas environment [150]. A more recent work by Parmar and Shin [151] found that fs laser creates globular micro/nano structures in air and elongated columnar structure in water, and both structures produce black silicon with wideband antireflection effect. Later a similar effect was achieved with a parallel microgroove pattern produced by fs laser [152]. The groove period is 100 μm and the width and depth are about 50 μm in size. The antireflection effect extends to the mid-IR wavelength range. Enhanced surface optical property is also demonstrated for metals through fs laser micro/nano structuring. A 95% absorptance for black platinum (Fig. 13a) over the wavelength range from UV to near-IR is achieved with laser induced micro/nano structures on the surface [114]. Yang et al. [153] reported significant enhancement of thermal emission to about 100% for a NiTi alloy due to a microscale coral-like surface structure produced by a fs laser. Surface textured silicon and metals have a wide range of potential applications including solar cells, detectors, sensors, field emission devices, plasmonics, broadband thermal sources, radiative heat transfer devices, etc.

Another unique capability of fs laser is to modify and control material's wetting properties. Superwetting surfaces were created using fs laser for silicon and human enamel and dentin tissues by Vorobyev and Guo [154, 155]. The structure consists of parallel microgrooves with a period of 100 μm with naturally formed micro/nano scale morphology over the microgrooves. When water is dropped on the processed area, it spreads rapidly along the microgroove orientation and can even climb up the surface vertically. A similar microgroove micro/nano hierarchical structure created on platinum, titanium and brass turned the surface superhydrophobic [156]. The wetting property of fs laser patterned surfaces is determined by the combined effect of surface morphology and surface chemistry. Skoulas et al. demonstrated the effect of surface morphology on wettability for a Ni film with different complex micro/nano structures as shown in Fig. 13b which were created by using a cylindrical vector fs laser beam [142]. Combining fs laser texturing and surface chemical treatment, the initial superhydrophilic metallic and dielectric surfaces after laser processing become superhydrophobic with contact angles exceeding 150º [157-159]. Materials with tailored wetting properties have potential applications in microfluidics, biomaterials, anti-icing, self-cleaning, etc. For example, Shashank and Shin developed a fast and inexpensive method to create microfluidic devices with textured superhydrophobic inner channel walls for controllable fluid flow [160]. A textured superhydrophobic surface created on a copper mold was transferred to the inner surface of a PDMS microchannel that increased the fluid flow rate by 186%. While the above mentioned studies concern surface wetting properties in air, a large body of work have been reported in the literature in recent years on fs laser induced underwater superoleophobicity. In a comprehensive review article, Yong et al. presented various fs laser structured surfaces that display underwater superoloephobic behaviors based on the design principle of "in-air superhydrophilicity to underwater superoleophobicity [161]. Potential industrial and military applications of superoloephobic surfaces were illustrated.



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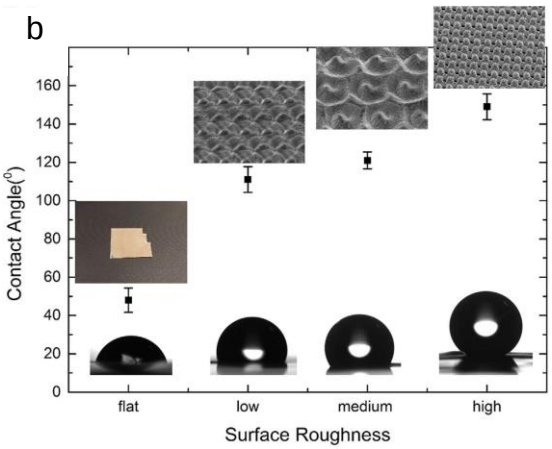

Fig. 13. Various fs laser functionalized surfaces (a) black platinum [114] and (b) wettability of Ni film with different structures (adapted from [142] on the basis of open access).

Femtosecond laser surface texturing for tribological applications has been an area of interest for almost 20 years. LIPSS has been generated and tested for friction and wear for various materials including metals, ceramics, and semiconductors. It has been shown that LIPSS can reduce friction under both regular sliding contact conditions [162] and harsh machining environment (i.e. chip-tool contact with high temperature, high pressure, and high speed) [163, 164], which is especially true under lubricated conditions. Concerning wear, however, LIPSS created on titanium was found to be ineffective under reciprocal sliding tests and the authors concluded that the LIPSS amplitude should be larger than the contact deformation depth for a beneficial effect [165]. In contrast, fs laser textured cutting tools with microholes or microgrooves of tens or hundreds of microns in size have been shown to be effective in reducing friction and wear under both dry and lubricated conditions in machining applications [166, 167]. This research area of textured tools has been active for about 10 years, with a continuous flow of publications targeting new textured tool development and different machining operations [168, 169].

4.3 Thin film machining

Femtosecond laser machining of thin film materials is driven by applications including thin film solar cells, MEMS, microelectronics, etc. In the production of a thin film solar device, a large solar panel is generally divided through laser scribing into narrow photovoltaic (PV) cells connected in series. Fig. 14 shows a typical structure of a thin film solar cell. The typical thickness is up to a few hundred nanometers for the two contact films and a few hundred nanometers to a few microns for the middle PV film, depending on the solar cell type. Scribing is a key manufacturing process that consists of three steps (commonly known as the P1, P2, and P3 scribes as shown in Fig. 14), with different film deposition processes in between. The P1 scribe removes the first thin film (bottom contact) from the substrate and defines the individual cells. The P2 scribe removes the absorber film to form the series interconnects between cells. The P3 scribe removes the second contact film (top contact) only or together with the PV layer to isolate each cell. The three scribes plus the gaps in between form the dead area width, i.e., the area that does not contribute to power generation. From the standpoint of cell performance, the scribes should be narrow with straight walls so that the dead area is minimized. Scribing should have high depth resolution so that only the selected film is removed. The scribes should be clean and free from damage. Comparative studies for ns, ps and fs laser scribing have shown that ns laser causes shunt between the top and bottom contact due to melting of the absorber film containing metallic elements [170]. In contrast, the benefits of fs and ps laser scribing are reduced thermal effects, less diffusion between layers, minimal cracking, and improved electrical performance [171]. However, sometimes a ps laser generates considerable melting as shown in Fig. 15 in P3 scribing of a CIGS solar cell [172]. This problem can be mitigated through careful design of process parameters via experiments and 3D two-temperature modeling (TTM) of the scribing process, as shown in Fig. 16 by the high quality P2 and P3 scribing on a CIGS solar cell achieved by Zhao and Shin [173, 174].

An alternative process known as direct material lift-off was investigated for various solar cell structures [175-179]. In the lift-off process, absorption of laser irradiation occurs mainly at the interface, which causes rapid, localized heating and plasma formation so that the top layer is ejected. Although undesirable for direct film ablation, ns laser was able to achieve successful P2 scribing from glass side film lift-off for a CdTe-based solar cell [179]. Krause et al. demonstrated that all three scribing steps were successful for a silicon based thin film solar cells [178]. Markauskas et al. achieved lift-off for P2 and P3 scribing with both fs and ps lasers for CZTSe thin film cells [177]. In contrast, no lift-off was realized for a CIGS based solar cell with both fs and ps laser at the wavelength of 1030 nm according to Gečys et al. [176], and the main reason is due to absorption throughout the thin film stack that lead to direct ablation. The dependence of the lift-off process on process conditions was demonstrated by Bayer et al. [175] in their study of perovskite film scribing. Three material removal mechanisms were identified as ablation only, lift-off, and ablation plus lift-off, as shown in Fig. 17. Depending on laser parameters and the irradiation method (e.g. film side or through glass), a specific mechanism may dominate the scribing process. An interesting technique named “rail-roading” patterning was presented by Jeoung et al. [180], in which two beams were focused on the surface with adjustable distance between the focal spots. When applied to scribing of a CIGS solar cell, it was found that the material removal displays features of both ablation and lift-off.

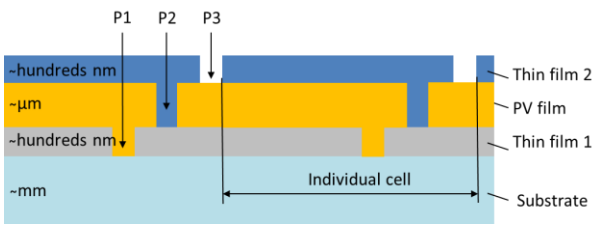

Fig. 14 Typical layered structure of thin film solar cell

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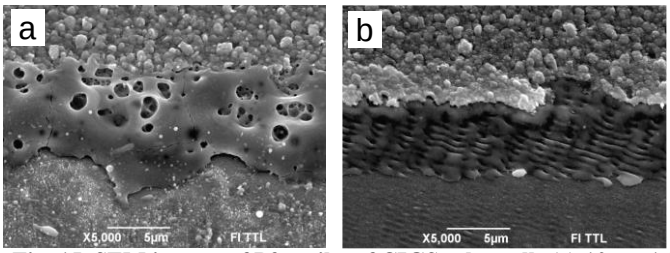

Fig. 15. SEM images of P3 scribe of CIGS solar cell: (a) 10 ps, 1.2 m/s and (b) 300 fs, 4 mm/s [172]. The scale bar is 5 μm.

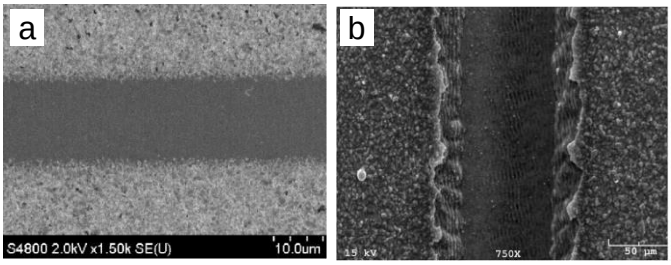

Fig. 16. SEM images of P2 and P3 scribing on CIGS [173, 174]: (a) P2: overlap ratio of 81 % and laser fluence of 3 J/cm^2 for ablation depth of 1.75 μm, (b) P3: laser fluence of 4.5 J/cm^2 and overlap ratio of 87% for ablation depth of 1.95 μm.

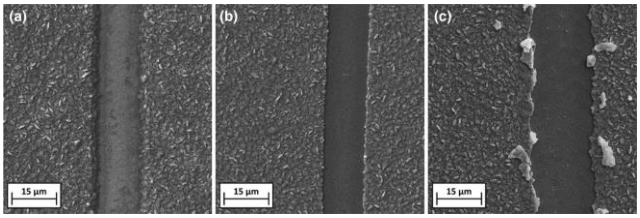

Fig. 17. SEM images of perovskite film scribing: (a) ablation, (b) ablation plus lift-off, and (c) lift-off [175]. The scale bar is 15 μm.

The desire for narrow scribes led to studies with tight focusing using microscope objective lenses. Clean, few micron P1 scribes were obtained for Mo on glass and ITO on glass [181-183]. Extension to P2 is challenging because of the much thicker absorber layer. To overcome the short depth of focus intrinsic to tight focusing without sacrificing for small spot size, Bessel beams were used for P1 and P3 scribing of Mo on PI and ITO on glass [184, 185]. The long central core of the zero-order Bessel beam is the useful part while the outside rings are undesirable because they may be strong enough to cause damage. To offset the detrimental ring effect of a Bessel beam, Yu et al. successfully constructed superposed Bessel beams using SLM to suppress ring damage observed with the zero-order Bessel beam [186]. One roadblock for industrial adoption of fs laser scribing is its low speed. Efforts have been made using fs lasers with a high repetition rate to reach scribing speeds on the order of m/s [187, 188]. It was shown for a CIGS based solar cell that all P1, P2 and P3 can be done at the speed of 2 m/s using one laser source but different laser fluence [188].

Most other research conducted on fs laser thin film machining has targeted various applications including MEMS [189], microelectronics [190], photonics [191], biosensors [192], electrochromic films [193], etc. 100 to 200 nm sized grooves and craters were created through direct ablation on metallic thin films without the use of high NA microscope objective lenses [194, 195]. The key is to have a highly stable laser beam and carefully controlled peak fluence to just a few percentage above the damage threshold, and a MHz level repetition rate seems to be beneficial for nanoscale machining. The effect of property mismatch at the interface for a Cr on glass substrate was brought out by Kim and Na [196], in which the glass serves as a thermal barrier for heat diffusion and induces a greater hydrodynamic flow and recoil pressure in the 500 nm thick film than the 200 nm film, resulting in different surface morphology. Besides the common method of single beam direct ablation, laser induced forward transfer was used to pattern a quartz substrate from a gold film donor plate [197]. Laser fluence had to be precisely controlled just above the threshold to enable efficient film transfer. Micromachining using a mask pattern was studied by Nakata et al. [198]. Microscale features in the form of lines and numbers were generated on a gold film with a single shot exposure that has potential for fast and large area patterning.

4.4 Machining in bulk of materials

4.4.1 Machining inside transparent materials

Ultrafast lasers possess unique capability of three dimensional (3D) processing inside transparent materials. When a tightly focused ultrashort laser pulse irradiates onto a transparent target (photon energy lower than the band gap), the extremely high laser intensity will initiate the multiphoton absorption, and excite electrons from valence band to conduction band. The material will be heated with a phase or structural modification, leading to a localized permanent property change. The occurrence of this process requires that the laser intensity is higher than the threshold of the multiphoton absorption, making it possible for localized 3D volume processing inside the sample by a tightly focused laser beam. Ultrafast laser processing inside transparent materials has been reviewed in a few recent publications [199-201]. Ultrafast laser irradiation induces phase and structural modification inside transparent materials. In this section, the mechanism of refractive index modification in different regimes is described and functional structures inside transparent materials fabricated by ultrafast lasers with a focus on femtosecond lasers are reviewed over the recent past.

(a) Types of refractive index modifications

The phase and structural changes according to the ultrafast laser parameters have been studied for various glasses such as porous glass [202], lithium niobium silicate glass [203], and silica fibers [204]. The ultrafast-laser-induced refractive index modification is categorized into three different regimes (or types): regime I (smooth refractive index changes), regime II (birefringent zones), regime III (micro-voids or damages) [205, 206]. Type I modification occurs due to a multiphoton absorption process with the pulse energy below the ionization threshold, which results in isotropic smooth changes in refractive index. Type II modification occurs due to avalanche ionization and the local production of moderate plasma, which results in ascribed either to laser-induced stress or self-ordered nano-gratings leading to anisotropic reflection. Type III modification occurs due to an excessive plasma production and subsequent Coulomb explosion, results in voids or disorganized

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damage features in the glass matrix [207, 208]. Each type of modification has the threshold depending on pulse duration and pulse energy. For example, as shown in Fig. 18, Type III modification is achieved inside the pure silica at high pulse energy (>300 nJ) and high pulse duration (>200fs).

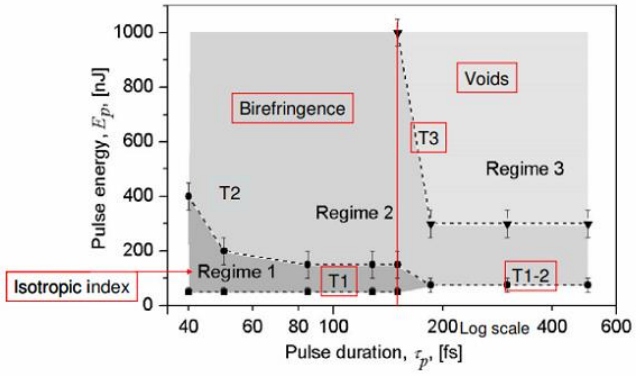

Fig. 18 Threshold pulse energies of a fs laser for different regimes of refractive index modification in fused silica (Adapted from Ref. [206] on the basis of open access)

(b) Waveguides

Refractive index changes in bulk materials by fs laser has been employed for buried optical waveguides fabrication with various geometries [209]. The fs laser processing can modify the refractive index with arbitrary 3D designs in a single process step inside any materials, even in photo-insensitive materials. Khalit et al. fabricated a series of 7 mm long straight waveguides inside a silver-containing zinc phosphate bulk glass by using a 1030 nm femtosecond laser with 390 fs pulses and 0.02 - 0.15 μJ pulse energy [210]. Morris et al. fabricated the channel waveguides in Tm:Lu_2O_3 ceramic by using a 1040 nm ultrafast laser with 200 fs pulses and 0.4 - 5 μJ pulse energy [211]. Patzold et al. fabricated the straight and s-curve waveguides in a PMMA foil using a 1048nm Yb:KYW laser with 600 fs pulses and 0.45 μJ pulse energy [212]. Consequently, the waveguides are usually fabricated by fs lasers with relatively low pulse energy as the type I modification.

(c) Bragg gratings

Bragg gratings are also inscribed inside the bulk materials by fs lasers. Bragg gratings is a periodic refractive index changes that perform as an optical filter that reflects a specific wavelength of light [213]. Femtosecond laser processing enables inscribing the gratings that are thermally stable up to the glass transition temperature in certain laser parameters [214]. It allows the ultrafast-laser-inscribed gratings to have a potential to be an element of the sensors for extreme conditions like high temperature [215, 216]. Femtosecond laser processing has been used to fabricate waveguides composed of volume Bragg gratings in bulk materials such as diamond [217] and PMMA [218].

In recent studies, fs lasers are used to fabricate fiber Bragg gratings (FBG) in various optical fibers such as a pure silica optical fiber [219], fluoride glass fibers [220], a cyclic transparent optical polymer fiber [221], TS-doped PMMA optical fibers [222], perfluorinated graded-index POFs [223]. Femtosecond laser processing for fabrication of FBGs with novel geometries have been studied [224, 225]. For example, as shown in Fig. 19, Ertorer et al. proposed damage-free beam delivery method using fs laser irradiation under oil-immersion [226]. Since the filament is generated due to Kerr-lens self-focusing effects, uniform gratings with high aspect ratio were inscribed inside the optical fiber.

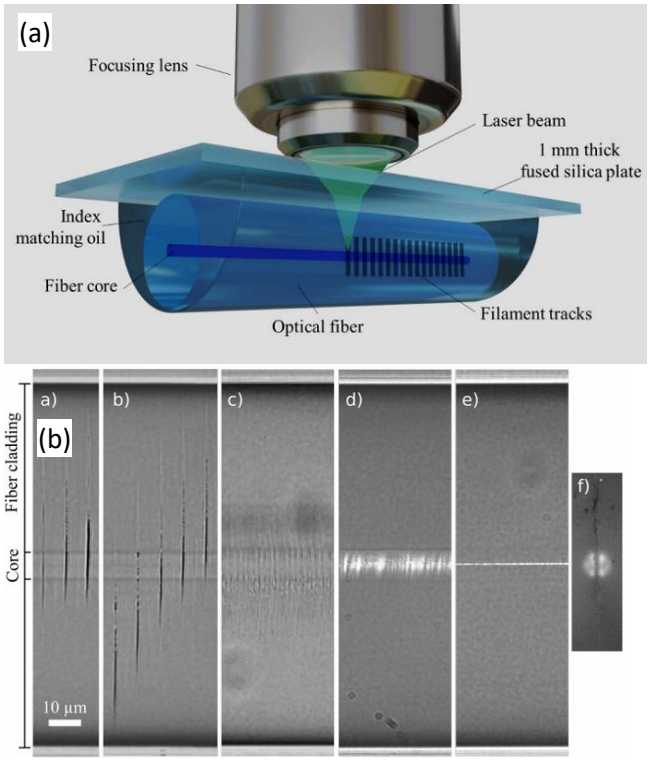

Fig. 19 (a) Schematic of the femtosecond laser writing of filament track Bragg gratings with oil immersion method (b) microscope images of isolated filamentation tracks (Adapted from Ref. [226] on the basis of open access)

(d) Microfluidic structures

Femtosecond lasers are also used as a 3D micromachining tool for fabrication of fluidic devices with two strategies: fs laser assisted wet chemical etching and liquid-assisted fs laser drilling [227, 228]. Femtosecond laser processing induces modification of chemical properties like etching rate [229], so that a wet etching process can be used to remove the irradiated region. Since the liquid like water assists to remove the debris during the laser processing, long and continuous channels can be fabricated inside the materials by the liquid-assisted fs laser drilling [86].

Recently, the fluidic devices with complex designs have been proposed to improve their integration and uniformity of micro-fluidic structures such as microfluidic channels, a microvalve, and a micropump in the microfluidics as shown in Fig. 20. Shan et al. proposed 3D multi-microchannel helical mixers fabricated by the fs laser assisted wet etching method. They performed two-step wet etching in order to obtain the hollow spiral channels with high uniformity [230]. Wang et al.

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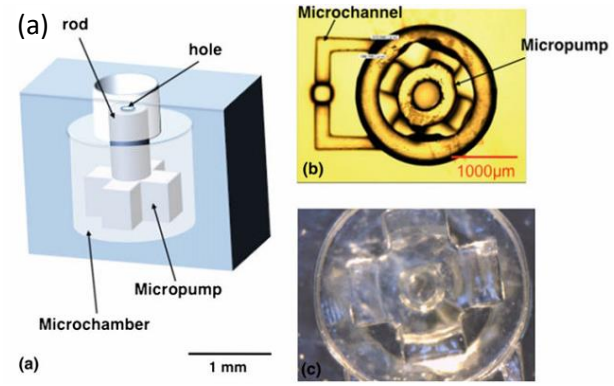
proposed a temporally shaped fs laser Bessel-beam-assisted chemical etching method to generate deep micro-channels in fused silica. The etching depth was enhanced by a factor of 13 compared to the conventional method because of the advantage of the Bessel beam such as ultralong focal depth, an intense central core, and uniform intensity distribution [231]. Accordingly, fabrication of microfluidic structures with high integration and uniformity are achieved with optimized conditions for the fs laser processing and etching process.

Polymer-based fluidic structures fabricated by fs laser have also been studied. Roth et al. proposed fs laser processing for 3D-micro-fluidic channels fabrication inside PMMA bulk material. They fabricated the micro-channels in two-steps: fs laser irradiation and annealing. Non-periodic disruptions inside the focal volume generated by the fs laser irradiation become continuous channels with cross section larger than the focal volume after annealing. The height, width and aspect ratio of the channels were adjusted by controlling laser conditions [232]. Vazques et al. also used fs laser in the fabrication process for an optofluidic cytometry platform based on PMMA. The fs laser was used as a welding tool for the PMMA parts [233].



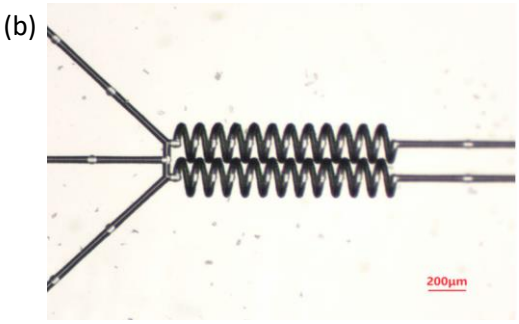


Fig. 20 (a) Micropump fabricated in 3D microfluidic structure in photosensitive glass and (b) Multi-Microchannel Helical Mixer Fabricated by femtosecond laser inside fused silica (Adapted from Ref. [227] with a permission and [230] on the basis of open access)

(e) Microscopic voids, nanocrystals and dichroic structure

Femtosecond laser can also induce formation of nanopores and voids inside the materials. A single void formation is caused by strong explosion and shock wave formation in decompaction regime. As shown in Fig. 21, the decompaction structure is observed for different laser scanning speeds. Ma et al. found that the periodic void structures can be controlled inside porous glasses by the mean pulse energy and the scanning speed [234]. Moreover, the ultra-short laser pulses induce microscopic voids in the direction of laser propagation due to self-focusing and filamentation, standing electron plasma wave or spherical aberration.

The fs laser processing with high repetition rate can create thermal field in the laser focused area, which results in space-selective precipitation of various functional nanocrystals inside glasses [235]. The nanocrystals inside the glasses can be used for photonic applications such as laser frequency conversion devices, 3D optical-data storage, optical grating, 3D multicolored industrial art objects, and ultrafast all-optical switches.

Femtosecond laser irradiation inside an Ag nanoparticle containing glass was shown by Podlipensky et al. to change the spherical nanoparticles to oval shapes oriented in the laser polarization direction [236]. Permanent 3D anisotropic structures were created with the use of multi-wavelength fs pulses, which displays dichroic functionality. These structures are erasable using thermal annealing and thus could have applications as optical 3D storage devices.

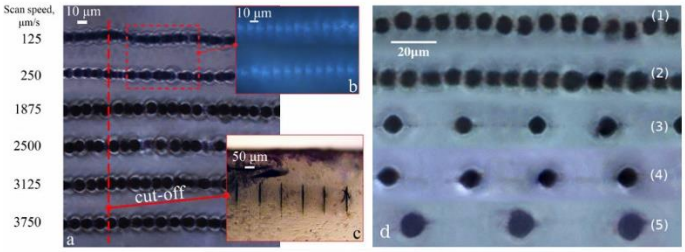


Fig. 21 (a-c) Optical images of decompaction regions obtained for different writing speeds; (d) image of periodic lines of voids formed inside of the porous glass (Adapted from Ref. [234] on the basis of open access)

(f) Color centers

Femtosecond laser processing for single crystal diamonds has increased in recent studies. Diamond has become a promising structure for spintronics and quantum information thanks to the remarkable properties of a nitrogen-vacancy (NV) impurity [237]. The Yb:KGW laser with 20-26 nJ pulse energy, 230 fs pulse duration, is focused inside the diamond using oil immersion lens. After laser processing, the diamond is annealed in a nitrogen atmosphere to produce the NV centers. It was found that the probability of creating single NVs to be 80% for the laser pulse energy of 24 nJ [238]. Hadden et al. proposed the waveguide-coupled NVs, which enables optically linked single NVs for single-photon sources or solid-state qubit in quantum photonic applications [239]. The intrinsic structure of the fs laser modified region and the model of phase transition including graphitization and photolytic formation of vacancies have been also investigated in references [240, 241].

4.4.2 Machining inside non-transparent materials

Ultrafast lasers can also be used to create structures inside materials that are opaque to visible light. This is possible because as long as the photon energy is below the bandgap,

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lasers beams can penetrate deep inside the material without significant linear absorption, and nonlinear light-matter interaction will initiate laser energy deposition at the beam focus. A commonly studied material that has scientific and technological significance is silicon, which has a bandgap of 1.1 eV at room temperature (corresponding to 1.1 μm laser wavelength in vacuum). A large body of work related to bulk processing in Si is stealth dicing of silicon wafers [242-246], which typically utilizes long-pulsed lasers to create internal defects followed by mechanical fracturing. When using femtosecond laser pulses for Si bulk processing, one needs to consider the high peak power and intensity that can create electronic and thermal nonequilibrium, which can in turn alter laser beam absorption and propagation. One can refer to the work by Verburg et al. who studied Si internal modification using 3.5 ns laser pulses at 1549 nm wavelength, and also gave a short review on Si bulk processing using short (ns) and ultrashort (fs-ps) laser pulses [247]. A notable application of bulk processing is writing waveguides, as demonstrated using nanosecond [248, 249] and femtosecond lasers [250, 251]. Nanostructures inside Si induced by 1.24 μm femtosecond laser pulses were studied by Mori et al. [252]. The orientation of laser-induced structures can be controlled by the polarizations of two laser pulses, and the periodic nanostructures may have the same origin as "ripples" that have been observed in dielectrics [253, 254]. In the studies by Grojo and coworkers [255], nonlinear absorption of Si at wavelengths longer than the linear absorption edge was found to pose a limit to the intensity that can be reached using femtosecond laser pulses. This is because nonlinear absorption taking place before the laser beam can reach its focus reduces laser energy and distorts beam profile. They also showed that this limit can be overcome by using solid-immersion focusing, which utilizes the high refractive index of silicon to increase the numerical aperture to ~3 [256]. Because of the transparency of silicon at wavelengths longer than 1.1 μm, ablation on the backside (beam exit surface) of silicon is possible, which has been demonstrated by several groups using fs-ns pulse durations [257-259]. Femtosecond laser beams with Gaussian and Bessel spatial profile have been used to investigate beam propagation in the nonlinear regime and to drill high-aspect-ratio channels in silicon [255, 260-262]. It has been shown recently that picosecond laser pulses can be used to create permanent internal modification in silicon, which offers an alternative to fabricating complex 3D structures [263].

4.5 Additive manufacturing using ultrafast lasers

In additive manufacturing that utilizes material melting, reflow and solidification, continuous-wave (CW) and long-pulsed lasers are often used to deposit thermal energy to initiate material phase change. 3D objects can also be additively manufactured using ultrafast lasers through linear or nonlinear laser-matter interaction. Recent advances in using ultrafast lasers for additive manufacturing are reviewed in the following sections.

4.5.1 Multiphoton polymerization

Lasers with ultraviolet (UV) wavelengths are commonly used in stereolithography, which utilizes linear absorption of organic molecules for UV light to induce cross linking of liquid photopolymers. Photopolymers typically consist of photoinitiators that will release reactive species upon UV laser radiation, and these species will initiate a chain reaction among monomers to form new bonds and cause solidification, thus "curing" the liquid photopolymer. This process requires short wavelengths (high photon energies) at which most photoinitiators are designed to have high absorption. It is possible to achieve photopolymerization through the simultaneous absorption of multiple, low-energy photons (multiphoton absorption), which is commonly referred to as multiphoton polymerization (MPP). One can refer to [264-270] for reviews of MPP and related applications. The most common form of MPP is two-photon polymerization (2PP) where two photons (typically visible to near-infrared wavelength) are absorbed simultaneously. However, three-photon polymerization is also possible [271]. Because the probability of simultaneously absorbing multiple photons is very small, in order for multiphoton absorption to take place, high peak laser intensities, typically on the order of 1 TW/cm^2 [272], are required. Such intensities can be provided by using ultrashort (<10 ps) laser pulses. 2PP has a few advantages compared to conventional single-photon stereolithography. Firstly, the laser beam can penetrate deep inside the photopolymer to initiate solidification only at the focal position, so the fabrication can in principle take place anywhere inside the photopolymer. This gives more freedom in the design of the fabrication process. Secondly, the fabrication resolution can be improved to the nanometer scale, because the material response scales with I^2, where I is laser intensity.

The principle of two-photon absorption was studied by Göppert-Mayer in 1931 [273], and was experimentally demonstrated by Kaiser and Garret in 1961 [274]. The idea of two-photon polymerization was proposed and demonstrated in early 1990s [275, 276]. In the concept proposed by Cabrera et al. [275], two laser beams cross each other in the photopolymer and 2PP occurs at the intersection. This concept has not been widely adopted because the structures that have been fabricated will obstruct the laser beams and cause distortion to the laser focus. Single-beam 2PP was realized by Strickler et al. [276], who used a mode-locked dye laser at 630 nm wavelength and a 1.4 N.A. (numerical aperture) oil immersion objective lens to fabricate structures as small as 100 nm. This small size was possible because of the quadratic intensity dependence of two-photon absorption, together with the fact that the features were fabricated nearly at the threshold and that some material could have been removed during rinsing. 3D spiral structures were fabricated by Maruo et al. [277]. In their work, a femtosecond laser with 200 fs pulse duration and 790 nm center wavelength was focused inside the photopolymer that was optimized in UV wavelengths. The quality and resolution of 2PP had been improved significantly since then, and the same group reported the fabrication of a micro-sized bull using 2PP that has the size of a red blood cell in 2001 [278]. They also showed that micro-sized mechanical devices with movable parts can be fabricated.

Photopolymerization, especially in the nonlinear light-matter interaction regime, is a complex process involving many micro- and macro-scale phenomena which have not been fully understood. Avalanche ionization, which can contribute to free carrier generation in dielectrics [279, 280], has found to have

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an influence in 2PP [281]. New insights into ultrafast dynamics in MPP can be obtained by studying optical nonlinearities [282] using, for example, the double-beam threshold measurement [283]. Spatial laser beam shaping is a powerful tool for MPP (and for laser material processing in general, see section 5.2 below). Multiple foci can be generated using diffracting optical elements and spatial light modulator to fabricate structures in parallel [284]. Non-Gaussian beams, such as non-diffracting Bessel beams, have been used [285] to fabricate high-aspect-ratio structures, and new optical devices can be used to generate non-Gaussian beams [286]. Materials that can be used in MPP have been expanded to have more functionalities, high resolution, and the ability to respond to external stimuli [287, 288]. MPP can be integrated with subtractive, laser machining processes for hybrid manufacturing [289].

4.5.2 Ultrafast laser additive manufacturing of metal, glass, ceramic, semiconductor and biological materials

The high peak power of ultrashort laser pulses can be utilized to achieve high temperature in materials to cause melting and solidification. Metal powders that have high melting temperature, such as tungsten, can be melted using femtosecond laser pulses at high repetition rate [290]. Copper has high thermal conductivity that prevents it from being additively manufactured using conventional lasers, but can be fabricated using ultrafast lasers [291]. This is because high temperatures can be reached and localized in the material. Rapid heating and cooling associated with ultrafast laser treatment will cause residual stress that will degrade the performance of the fabricated part. It remains to be seen how to improve the quality of ultrafast laser additively manufactured parts. Femtosecond lasers can also be used to melt glass powders [292], where nonlinear absorption is likely to contribute to the energy absorption. Applying the above-mentioned MPP technique, polymer-ceramic hybrid materials can be used to fabricate micro-needles for medical applications [293]. It is worth pointing out that ultrafast lasers can also be used for post processing of additively manufactured metal parts [294]. Figure 22 shows some examples of additively manufactured parts using ultrafast lasers.

In the above-mentioned additive manufacturing methods, heating and chemical reaction that are caused by laser energy deposition are used to directly introduce material change. Another approach to building 3D parts is to deposit or transfer materials that are more or less in their original state. This is best known as pulse laser deposition (PLD) [295] and laser-induced forward transfer (LIFT) [296]. These methods have enabled additive manufacturing of semiconductor and biological materials.

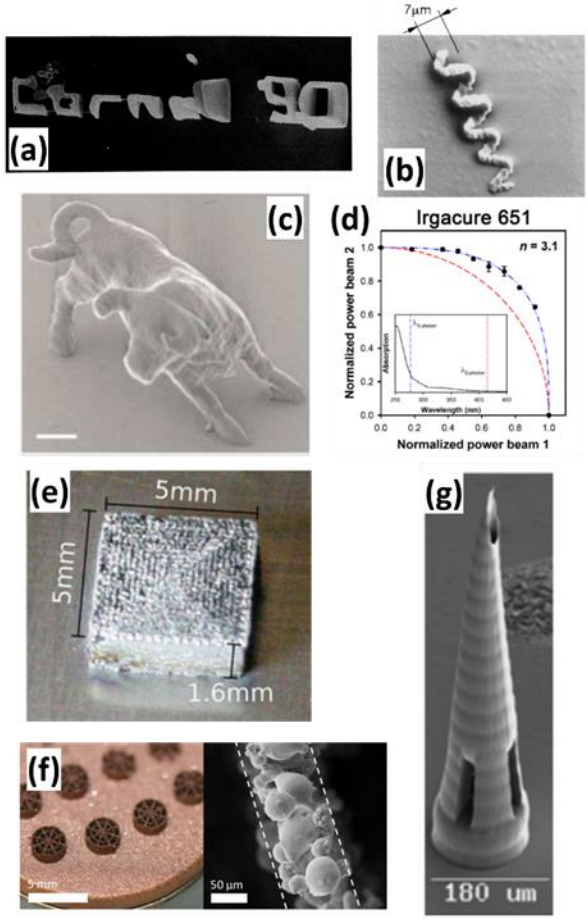

Fig. 22 Examples of additive manufacturing using ultrafast lasers. (a) Early work of microfabrication using two-photon polymerization (2PP) [276]. (b) 3D spiral structures fabricated by 2PP [277]. (c) A micro-sized bull sculpture fabricated by 2PP. Scale bar 2 μm [278]. (d) Determining the nonlinear absorption order (*n*) using the 2-beam initiation threshold (2-BIT) method [283]. (e) A tungsten cube additively manufactured by high-repetition-rate femtosecond laser pulses [290]. (f) Thin-wall structures made of copper powder is built by a femtosecond laser. An SEM image of the microstructure is shown on the right [291]. (g) A micro-needle is printed using 2PP in polymer–ceramic hybrid materials [293].

4.6 Ultrafast lasers in biomedical applications

In the past two decades ultrafast lasers have become a mainstream in biomedical applications, and found their place in clinical practice. The ability to alter biological media on subcellular level without disrupting cell viability highlights the use of ultrafast lasers in research, whereas, high precision and modest heat affected zone are primary factors in adopting these lasers in clinical applications, especially ophthalmology. Furthermore, the latest research demonstrates the ability to enhance material properties of collagen rich tissues without disruption, which opens a completely new avenue for applications and may in the future transfer to manufacturing as well, perhaps as means for inorganic polymer treatment.

Non-linear absorption, a unique property of the ultrafast laser, enables treatment of the interior of target material without affecting its outside surface. Proper use of optical and laser

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parameters provides control over different processing regimes, photodisruption (PT) and ablation via thermal accumulation (ATA), both of which are known processes in nontraditional manufacturing. PT is an underlying mechanism in femtosecond laser-assisted corneal refractive surgeries, an application already mature in clinical practice. Amplified pulse trains at few kHz repetition rate with pulse energies above the damage threshold are used for bubble formation [297-301], which plays a key role in the surgery process. Cuts are created by the superposition of a series of cavitation bubbles.

Nanoscale femtosecond applications have been present for about a decade, and can be broadly characterized by use of long series of pulses from femtosecond laser oscillators with repetition rates on the order of 80 MHz and pulse energies around the optical breakdown threshold [302-305]. Stuart and co-workers have been developing theoretical framework for nanoscale applications of femtosecond lasers in biological media [306-308]. Most reported studies focus on a sub-cellular level for cell and tissue studies [309, 310]. A number of researchers have focused on the nanosurgeries in which ablation is used to remove cellular components without affecting the viability of the cell itself (Fig. 23). Konig and co-workers have been performing nanodissection of human chromosomes with femtosecond laser pulses [301, 311, 312]. Tirlapur and co-workers [303] utilized ultrafast lasers for generation of reactive oxygen species in mammalian cells to induce their apoptosis-like death. At the moment there are few prospective clinical applications with one being removal of the small content of biomaterial for very precise subsequent proteomic or genomic analysis [313].

Advantages of using oscillator systems in biological applications are numerous, including increased resolution, reduced trauma to surrounding tissue due to smaller total energy deposition and affordability of oscillator systems in comparison to the chirped amplified systems capable of producing pulses with significantly higher energy.

Femtosecond lasers are used in vision correction surgeries, anterior lamellar corneal transplantation, keratomileusis, channel creation for corneal implants [314], as well as cataract surgeries. Laser in situ keratomileusis (LASIK) is a vision correction surgery comprised of flap formation via a mechanical microtome followed by the photoablation of the stromal tissue with an excimer laser. The femtosecond laser was introduced to replace the mechanical microkeratome to create a flap in the so-called Femtosecond LASIK [315]. In the next generation surgery called small incision lenticule extraction (ReLEx smile) [316-318], a femtosecond laser alone is used to reshape the cornea by directly creating channels within its interior without removing the epithelium, the outermost layer of the cornea.

Juhasz and coworkers [319] conducted initial studies on use of femtosecond lasers in laser assisted corneal surgery in the mid-1990s. They used water to model shock wave generation and subsequent cavity formation and applied the same method to bovine corneas. Further, it has been shown that femtosecond generated flaps have numerous advantages in comparison to mechanical keratomes including decreased risk of infection and reduced intraocular pressure during the procedure, greater precision and more uniform thickness [315, 320]. Lubatschowski has investigated the differences in laser-tissue interaction and beam delivery of different commercial systems tailored for refractive surgery [321]. Repetition rate effects on affected tissue have also been studied [322]. Significant differences in the nature of cell death as a function of laser repetition rate have been observed. At lower repetition rates the laser pulse energy required to create a cut is higher resulting in predominant cell death by necrosis rather than apoptosis. As necrosis causes release of intracellular contents into the surrounding tissue, there is more inflammatory cell influx after flap formation. Revealing the cavitation bubble formation mechanisms at different processing and beam delivery regimes will help to optimize laser parameters, control the creation of bubbles, and reduce complications.

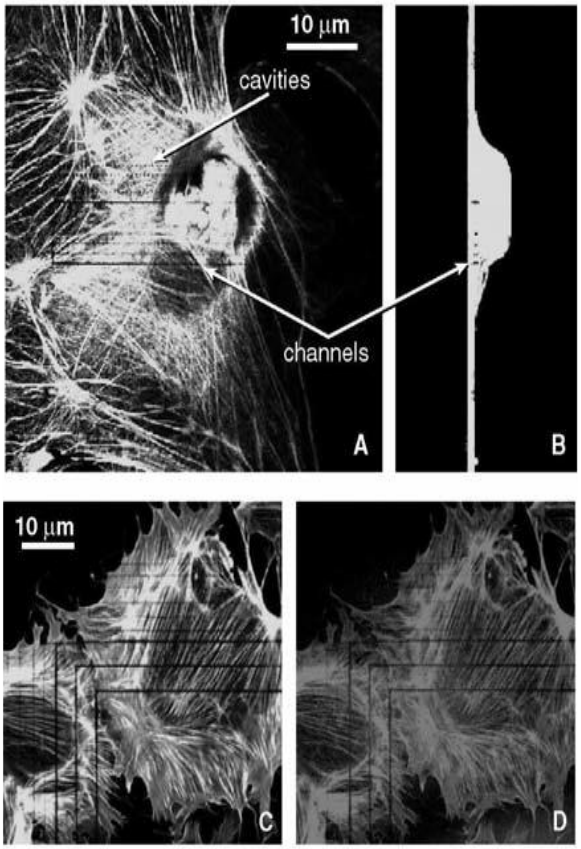

Fig. 23 [300] Laser confocal microscopic images of a fixed 3T3 fibroblast stained for F-actin with a green fluorescent dye. (A) Top view of a mid-plane horizontal section through the cell showing channels and cavities produced by femtosecond laser ablation. The top and bottom channel were obtained at a pulse energy of 3 nJ; those in between at pulse energies ranging from 1.5 to 2.3 nJ. (B) Reconstructed orthogonal image of the same cell. (C) A cell that was prestained for F-actin with a green fluorescent dye immediately after ablation with 2-nJ (wide channels) and 1.5-nJ (narrow channels) laser pulses. (D) The same irradiated cell after restaining for F-actin with a red fluorescent dye.

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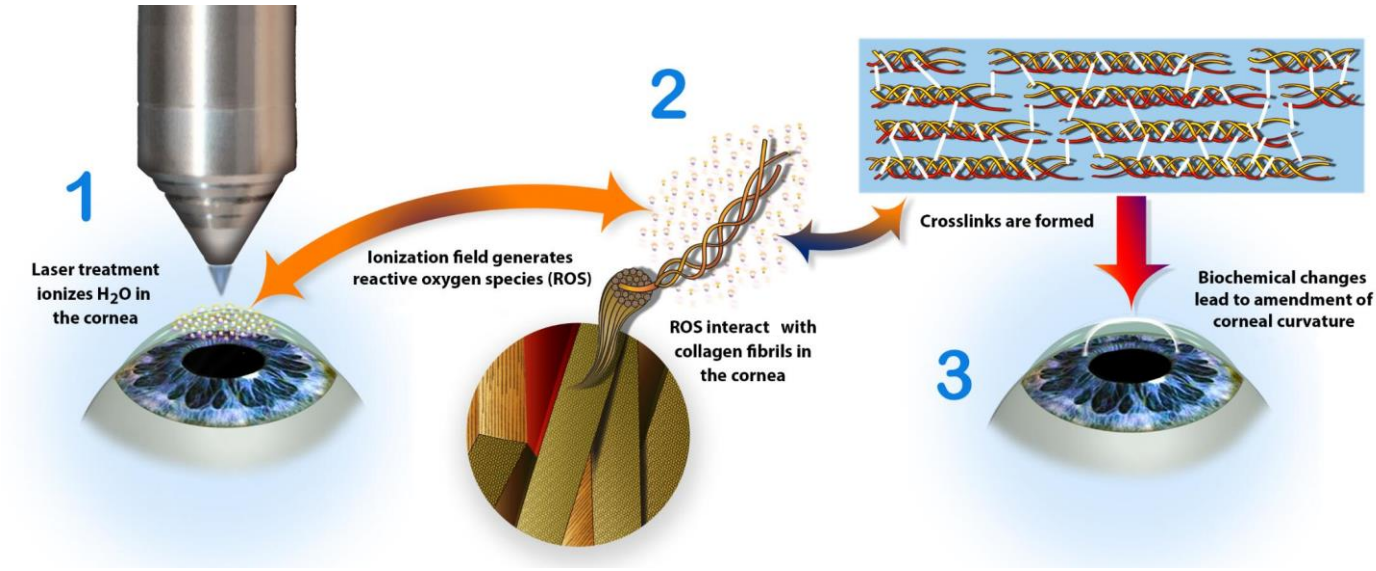
1
Laser treatment ionizes H_2O in the cornea
Ionization field generates reactive oxygen species (ROS)
2
ROS interact with collagen fibrils in the cornea
Crosslinks are formed
Biochemical changes lead to amendment of corneal curvature
3

Fig. 24 [323] Step-by-step illustration of the use of the proposed laser-assisted process for the non-invasive correction of refractive errors.

Recently, Vukelic and co-workers introduced a novel ultrafast laser-tissue interaction regime that relies on photochemical effect induced by ultrashort laser pulses rather than tissue disruption [323, 324]. The approach restricts the laser regime such that the treatment is always below the optical breakdown level, and thus relies on the ionization potential for alteration of corneal geometry (Fig. 24). If a femtosecond laser operates below the energy level required for optical breakdown, ionization of atoms within the focal volume is still possible, because the ionization probability has a number of resonance maxima due to intermediate transition of the atom to an excited state. In the vicinity of such maximum, the ionization cross-section increases by several orders of magnitude enabling ionization even if the frequency of the incoming electromagnetic wave is lower than the ionization potential. Such a scenario enables formation of an ionization field, which locally ionizes and dissociates water content in the tissue, creating reactive oxygen species (ROS). When applied to collagenous tissues, the reaction between ROS and surrounding proteins results in crosslink formation (Fig. 24), giving rise to spatially resolved alterations in mechanical properties. Carefully tailored alterations of mechanical properties of collagen stroma can be utilized to change the overall corneal curvature that may be leveraged for the correction of refractive errors. Another explored application is strengthening of articular cartilage towards addressing early osteoarthritis [325, 326].

4.7 Femtosecond laser-based super-resolution manufacturing

The holy grail of nanoscience is to manufacture functional structures from the atomic scale to the macroscopic in three dimensions. The minimum size attainable by conventional laser direct writing is limited by Abbe's diffraction limit,

$$d \approx \frac{\lambda}{2n \sin \alpha} \quad (3)$$

where d is the minimum beam diameter, λ the wavelength, n the refractive index of the medium where the light is focused, and α the beam divergence angle. Current semiconductor chips are produced by photo-lithography with continuous shrinking of wavelength from UV, deep UV to even extreme UV. The required mask masters are fabricated using e-beam lithography subject to the shorter de Broglie wavelength of a focused electron beam. This approach leads to nanomanufacturing equipment capital cost to reach the $100M level, thereby forming an economic barrier for extensive applications of nanodevices [327, 328]. Based on the interaction between ultra-fast laser and materials, there are two basic methods to overcome the optical diffraction limit: near-field optical manufacturing and innovative far-field nanolithography. Near-field optical manufacturing is intrinsically limited to the surface, i.e., to 2D, planar lithography [329] which can be conducted with either metallic tip-assisted laser processing or near scanning optical microscopy [330, 331]. The key to realizing 3D nanolithography is to focus the pulsed laser into a diffraction-limited spot or a depletion region limited by a donut-shaped, stimulated light in a photon-sensitive medium [332].

A nonlinear multiphoton absorption procedure, such as two-photon polymerization (2PP), can enable precise manufacturing at a resolution near the diffraction limit [333-335]. The probability of the multiphoton absorption effect, p_n, is a power function of laser intensity I as follows [336],

$$p_n = \delta_n I^n \quad (4)$$

where δ_n is the absorption cross-section of n photons, which is dependent on the nth nonlinear coefficient of the optical medium. Therefore, the multiphoton effect can only occur at an extremely strong intensity, e.g. the central regime of a focused

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laser beam. As shown in Fig. 25(a), 2PP can be controlled by the laser intensity. When I is higher than I_o, the threshold, 2PP will occur at the central area. Theoretically, the size of 2PP is about $\lambda/(2.7n\cdot\sin\alpha)$, significantly smaller than the diffraction limit of $\lambda/(2n\cdot\sin\alpha)$ by single-photon absorption [332].

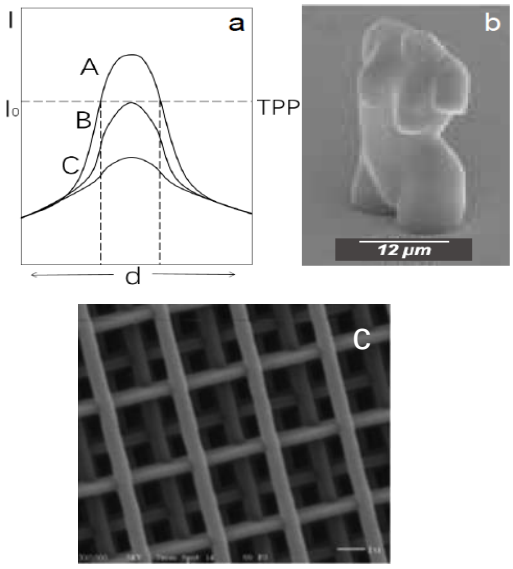

Fig. 25 (a) Gaussian-beam intensity for 2PP, (b) printed Venus micro-statue, (c) a printed woodpile-style photonic crystal. Fig. 25 (b, c) are adapted from Ref. [334, 335].

Serbin reports 2PP direct writing with tightly-focused femtosecond laser pulses (60 fs, 90 MHz, 780 nm) using Ormocers as the photosensitive resin and a high numerical aperture, oil-immersed objective lens [334]. Fig. 25(b) shows a 3D printed Venus micro-statue and Fig. 25 (c) presents a woodpile-style photonic crystal [335]. The woodpile structure of photonic crystal in Fig. 25 (c) possesses a rod diameter of 300 nm and rod spacing of 900 nm. Although a photonic band gap is evident at the near-IR band in the photonic crystal, the crystal does not possess a complete photonic band structure due to fabrication imperfections.

Super-resolution manufacturing beyond the diffraction limit has been achieved by a Stimulated Emission Depletion (STED) mechanism inside a photon-sensitive polymer. An insoluble, photo-induced, cross-linked polymerization can be used for super-resolution manufacturing. There are three primary methods to realize a super-resolution depletion-limited focal spot. With STED lithography [337, 338], monomer molecules are first excited via two-photon absorption. The excited molecules are brought back to the ground state by the ring-shaped, stimulated light at a short wavelength via a stimulated emission. Therefore, only surviving radicals in the central regime can undergo polymerization. Alternatively, resolution augmentation through photo-induced deactivation (RAPID) lithography, excites photo-initiator (PI) molecules via two-photon absorption, generating a long-lived intermediate state. Upon further ring-shaped light excitation, the intermediate state is activated and does not lead to cross-linking polymerization [339]. Finally, two-color photo-initiation/inhibition (2PII) lithography [340] excites PI molecules via *one-photon* excitation and generated radicals. The photo-inhibitor molecules are excited by another ring-shaped light at a different wavelength via one-photon absorption, generating non-initiating radicals, which can terminate the polymerization of excited radicals. In summary, all these methods involve photo-sensitive polymers and realize a depletion-limited focusing through a spatially-controlled ultrafast photochemistry.

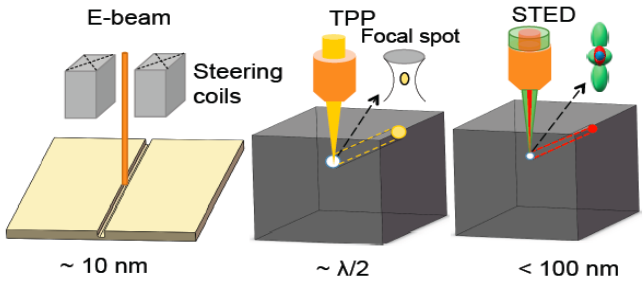

Fig. 26 Comparison of e-beam, 2PP and STED lithography. These figures are modified from Ref. [341].

In a typical STED configuration, the diameter of the exposure area can be calculated by the formula [342, 343]

$$d \approx \frac{\lambda}{2nsin\alpha\sqrt{1+bI_{STED}/I_S}} \qquad (5)$$

where $nsin\alpha$ is the numerical aperture, the same as in Eq. (3), b is a systematic coefficient dependent on the beam shape, I_{STED} is the intensity of the depletion light, and I_s is the switching intensity at which half of radicals will be depleted through a stimulated emission. It is apparent that one can reduce the exposure area by increasing I_{STED}/I_S ratios. Fig. 26 shows a comparison among the e-beam, 2PP, and STED lithography approaches. Although e-beam lithography can reach a resolution around 10 nm, it is still a near-planar manufacturing (2.5 dimensional) technique. Additionally, it requires operations to be accomplished in an ultrahigh vacuum chamber. 2PP is a facile 3D manufacturing technique with resolution close to the diffraction limit. STED is an ultra-precise 3D manufacturing beyond the diffraction limit.

Gan has demonstrated deep sub-diffraction optical beam lithography with a 9 nm feature size based on the STED mechanism [341]. They chose 2,5-bis(p-dimethylaminocinn amylidene)-cyclo-pentanone (BDCC) as the two-photon initiator and the monomer was SR399 or SR444. A 800nm 140 fs 80 MHz laser was used as the writing beam and the depletion light was a 375 nm CW laser. The donut-shaped light was generated through a planar vortex phase plate. Two-line resolution of 52 nm has been achieved at a depletion intensity of 2.42 μW/cm^2. Shown in Fig. 27, micro-scale optical cloaking has also been demonstrated with a specially designed photonic crystal with lattice varying from 500 nm to 900 nm [344].

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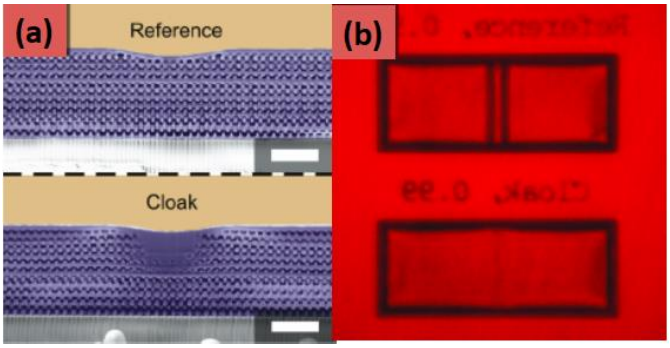

Fig. 27 STED-direct writing of photonic crystal and 3D optical cloak. (a) Photonic crystals (reference) with a lattice of 500 nm and a cloak with a lattice ranging from 500 nm to 900 nm, the bar is 2 μm; (b) Top-view of two golden lines under photonic crystal (above) and a cloak (below). Note that the lines disappeared beneath the optical cloak. These images are adapted from Ref. [344].

Although STED is a well-established method for overcoming the diffraction limit, there has been limited progress in direct writing of nanostructures using different materials other than polymers. Cao reported gold nanoparticles generated by using ion-doped hybrid photoresin [345]. Frolich reported a titania woodpile-style photonic crystal with a complete photonic bandgap via STED direct writing and a double-inversion procedure [346]. If a metallic nanostructure can be fabricated via STED direct writing, the super-resolution manufacturing can pave the way to various nanodevices for energy, environment and biomedical applications.

4.8 Numerical methods

The ultrafast laser has demonstrated its superiority for material processing and manufacturing. To further explore its application, it is critical to understand the ultrafast laser-matter interaction, which is a complex multi-physics process. In general, when materials are irradiated by an ultrashort laser pulse, the laser beam energy is first absorbed by free electrons (metals) or bonded electrons (semiconductors, dielectrics). By absorbing the incident laser beam energy, the electrons inside the target are heated to an extremely high temperature in a very short amount of time. The electrons will transfer the energy to the lattice, leading to phase change, structural modification, and ablation as described in section 4.3.1. The ejected material will be partially or fully ionized to form plasma. On the other hand, when the electrons near the target surface gain enough energy, surface electron emission (photoelectric or thermionic) will occur [347-349]. The ejected hot electrons are energetic enough to ionize the ambient gas, forming an early stage plasma in several or tens of femtoseconds [350, 351]. Due to the complexity of the process, numerical modeling is demanded to elucidate the underlying mechanisms. Extensive studies have been devoted to investigating the physics of ultrafast laser-matter interaction, and multiple useful models have been developed.

The two-temperature model (TTM) has been widely used to describe the energy transfer inside the target during ultrafast laser ablation. The basic governing equations are:

$$\frac{\partial}{\partial t}\left(C_e T_e\right) = \nabla \cdot \left(K_e \nabla T_e\right) - G\left(T_e - T_l\right) + S \qquad (6)$$

$$\frac{\partial}{\partial t}\left(C_l T_l\right) = \nabla \cdot \left(K_l \nabla T_l\right) + G\left(T_e - T_l\right) \qquad (7)$$

where t is the time, G is the electron-phonon coupling factor, S is the laser source term, C_e/C_l and K_e/K_l are the thermal capacities and thermal conductivities of electrons and lattice, respectively. The electrons and the lattice are treated as two separate systems with energy coupling between them. This approach is based on the fact that laser energy is first absorbed by electrons in the material, and then the electrons are relaxed by coupling with phonons. Anisimov et al. proposed the parabolic TTM [352], which can be generally used for pulses below 10 ps. However, it is not applicable for pulses shorter than 100 fs, because laser-plasma/plume interaction is not considered, and thermal equilibrium in the electron system is assumed for all time. Hyperbolic TTM [353, 354] was then developed for pulse duration shorter than 100 fs, considering electron non-thermalization. To further improve TTM, a semi-classical TTM was established by Chen et al. [355] to include the effects of non-equilibrium electron transportation and electron drifting. TTM was further combined with rate equations to consider the electron number density evolution during laser ablation of dielectrics and semiconductors [356]. One requirement for accurate prediction of ablation depth with TTM for a wide range of materials is to consider the correct thermal properties such as C_e and k_e as well as the electron-phonon coupling factor at very high temperatures. Wu and Shin used the quotidian equation of state (QEOS) and the model of Lee and More with TTM and demonstrated the efficacy in accurately predicting ablation depth of various metals [357] and semiconductors and dielectrics [358] over a wide range of fluence and laser pulse durations.

One issue with the commonly used TTM is that the predicted ablation depth is not deterministic at high fluence because the model does not consider heat loss by material ejection. In references [359, 360], TTM was improved to solve this issue by considering the energy loss through a dynamic description of the ablation process with material removal. TTM has its advantages in simplicity, computational efficiency, and decent prediction accuracy. However, it is a pure thermal model, which cannot describe the non-thermal processes (Coulomb explosion, laser-induced shock wave, etc.), plasma generation and evolution, and laser-plasma interaction.

Hydrodynamic (HD) models describe the evolution of the irradiated materials, with considerations of conservations of mass, momentum and energy on the continuum level [361]. HD models have been developed to describe the material removal and plasma expansion behavior during laser ablation [34, 362-367]. It has been widely applied to longer pulse ablation processes and is proved to be valid for various processing conditions [365, 368]. However, for ultrafast laser ablation, the surface hot electron emission and resultant early stage plasma generation are very important and need to be integrated into the HD model. In the model developed by Bulgakova et al. [369], the effect of hot electron emission process was accounted for and the electron transport inside the target was calculated. Chen and Mao applied a one-dimensional HD model to simulate the

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early stage plasma behavior during femtosecond laser ablation [370]. Colombier et al. incorporated TTM into their hydrodynamic model to account for the electron-ion coupling effect and thus more accurate simulation results on laser ablation were obtained [364]. Zhao and Shin developed a multi-fluid HD model to capture the dynamics of electrons, ions, and atoms separately [40, 350], and successfully simulated both early plasma and plume plasma. The application of HD model is limited by its disadvantages in the following aspects. First, equations of state (EOS) have to be combined with the HD model to provide thermal and optical properties of materials in a wide range of temperature and density. Therefore, the accuracy of a HD model strongly relies on the accuracy of EOS, which is not satisfactory in many cases. Second, as a continuum model, a HD model is not capable of capturing the non-equilibrium transformation and other ultrafast dynamics during the initial stage of ablation.

The molecular dynamics (MD) methods have been developed owing to the advancement of knowledge in solid state physics and quantum mechanics. Once the intermolecular potential is given, MD can be used to simulate many problems with their specific external forces following the Newton's Second Law. MD provides an explicit atomistic representation of target heating, material removal and expansion, and solves problems that cannot be accounted for by continuum models, such as highly non-equilibrium states and fast phase transformations induced by ultrashort laser pulse irradiation [371]. It has been widely used to investigate the ultrashort laser pulse ablation of different materials in different fluence ranges [22, 24-26, 372, 373], providing insight into the material removal mechanisms and fast transitions in the very early stage. MD model itself does not consider the electron relaxation dynamics. To improve it, Schäfer et al. established TTM-MD models by combining MD and TTM models together [374]. The thermal evolution of the electron system and its coupling with the lattice are involved in these models. In references [375, 376], an alternative improvement to MD models was proposed by coupling it with a Monte Carlo (MC) method, which was adopted to describe the electron dynamics during ablation. The ionization process and fast phase change of the electrons during ablation were described by the MC method, and the simulation results were improved. Inogamov et al. used an integrated model by a HD model and a MD model, where the HD model was applied to solve the early stage heating and expansion of electrons/lattice, and the MD model was adopted to describe the lattice atom motion and removal process later [377]. Hu et al. improved the MC-MD model by adding the particle in cell (PIC) method to calculate the Coulomb interaction between charged particles and the beam propagation method (BPM) to handle the interaction of early plasma with the incident laser beam [45]. As a result, the early plasma dynamics could be simulated with good accuracy in comparison with experimental measurements. This MD/MC-PIC-BPM model was further combined with an HD model in [42], where the output of the MD/MC-PIC-BPM model served as the initial stage of the HD model, and thus the plume plasma evolution in a longer time scale up to tens of ns could be covered. MD models have their own disadvantages in its computational cost. As a consequence, it can only simulate the ablation process in a very small simulation domain and time scale, which are not sufficient to capture the ablation process of the bulk material.

5. RECENT DEVELOPMENT IN ULTRAFAST LASER BASED MANUFACTURING

5.1 Mechanism of ultrafast laser induced damage studied by the pump-probe technique

For any laser-based processing technologies, it is crucial to have a fundamental understanding of how ultrashort laser pulses interact with materials, and what the key parameters are that influence the laser processing outcome. This is particularly important for ultrafast laser material processing, which involves light-matter interaction under extreme conditions. This section will review recent studies on the mechanism of ultrafast laser induced damage of materials, with a focus on obtaining time-resolved information of ultrafast laser induced material dynamics. Reviews of ultrafast laser – matter interaction at and above the damage threshold can be found elsewhere [378-380].

A useful tool for the study of ultrafast laser – solid interaction is the pump-probe technique, which is an in-situ, nondestructive measurement of laser induced phenomena in materials. In a typical pump-probe setup, a strong laser pulse ("pump") induces disturbance to the material, and a weak pulse ("probe") interacts with the disturbance whose information will be "encoded" in the probe pulse. By analyzing the transmission, reflection, phase, or a combination of these optical constants of the probe pulse as a function of the delay between the pump and the probe pulse, time-resolved information about laser-induced changes can be extracted. With ultrashort laser pulses, the temporal resolution of this technique is suitable for studying fast dynamics such as electron ionization and relaxation, laser-induced plasma, and shock waves. This technique has been used by many researchers in recent studies of laser induced damage and modification in solid materials. Mouskeftaras et al. used a pump-probe interferometry technique to measure the change of refractive index with 100 fs temporal resolution [381]. Their results suggest that avalanche ionization does not play a dominant role for ultrashort pulse irradiation. Mauclair et al. used pump-probe optical transmission microscopy and phase contract microscopy to study ultrashort pulse induced positive and negative refractive index change in fused silica and borosilicate glass [382]. Fast and slow material responses were observed, indicating the effect of carrier dynamics and material phase change. Pump-probe shadowgraphs were recorded in a study by Zhao and Shin on the dynamics of early plasma generation in femtosecond laser ablation of silicon [30]. Coulomb explosion caused by electron emission was found to increase material ablation rate. Using a train of two femtosecond laser pulses, Zhang et al. showed that nanostructures originated from electron dynamics can be controlled by changing the delay between the two pulses [383]. These nanostructures can be used in surface-enhanced Raman scattering. Garcia-Lechuga et al. observed an interesting ring pattern in pump-probe time-resolved microscopy [384, 385]. These patterns resemble Newton's rings, and are caused by light interference between an expanding layer of ablated material and the interface of a non-ablated substrate [385]. Similar patterns were observed by Rapp et al. in femtosecond laser ablation of silicon nitride [386]. Wædegaard et al. studied

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the evolution from free carrier generation to material ablation in single-crystal sapphire using pump-probe spectral interferometry and reflectivity measurement [387]. Pump-probe spectroscopy was used by Acharya et al. to study ultrafast carrier dynamics in ZnO and $BaTiO_3$ thin films [388]. The presence of excitons was observed, and excitons can be excited to inject carriers in hetero-structures. These experimental studies can often be used in conjunction with modelling and simulation [389-391] to reveal how ultrashort laser pulses interact with materials in the above-threshold regime.

5.2 Spatial beam shaping and temporal pulse shaping for ultrafast laser material processing

Material processing using ultrafast lasers is most commonly conducted using laser pulses at kHz – MHz repetition rate, with Gaussian spatial beam profile, and at near-infrared fundamental wavelength. These conditions are used because they are the laser parameters from most commercially-available ultrafast laser systems. However, for material processing applications, the optimal processing parameter set often varies from material to material, and even with the same material, different processing modes (drilling, cutting, surface texturing, etc.) require the tuning of processing conditions. This section is focused on recent advances in using temporal pulse shaping and spatial beam shaping techniques to tailor ultrafast laser material processing for improved manufacturing outcomes.

5.2.1 Temporal pulse shaping

Temporal pulse shaping can be achieved by a variety of techniques, and the most commonly used methods in laser material processing are Fourier synthesis of spectrally modulated waveforms, and the interferometric method based on Michelson and Mach-Zehnder interferometers [392, 393]. Li et al. adopted the interferometric method to create a pulse pair, which was used to determine the damage threshold as a function of the delay between the two pulses in dielectric materials [394]. Their work showed that the decay of free carriers must be taken into account when analyzing the laser damage threshold measurement. Choi et al. used three laser beams (two pump beams and one probe beam) to investigate the dynamics of plasma and shock wave in femtosecond laser ablation of crystalline silicon [395]. An increase of ablation volume was observed when the two pump pulses are at ~10 ps delay, and the increase was due to a better energy coupling to the sample, because at short delays (1-5 ps), liquid film or hot surface plasma formed by the first pulse will increase the reflection of the second pulse, while at longer delays (> 10 ps), the ejected material will reduce the transmission of the second pulse. The work by Qi et al. on femtosecond laser ablation of silicon showed that the energy coupling is enhanced at long delay because the density of the plasma formed by the first pulse will decrease, allowing deep penetration of the second pulse [396]. In a study with fused silica, Chowdhury et al. noticed that the depth of the laser ablated hole is maximum when two pulses are overlapped at zero delay, and the depth decreased as the delay increases. This is explained as the screening by the plasma produced by the first pulse [397]. The effect of femtosecond laser pulse train processing was investigated theoretically by Zhang et al. [398]. It was found that photoionization of self-trapped excitons (STEs) is important for understanding femtosecond laser ablation and controlling laser processing results. A pulse train consisting of four femtosecond laser pulses with varying delays was generated in a 2-fold Michelson interferometer, and this pulse train was used to process metal, semiconductor and dielectric samples, which were found to require the optimization of delays according to the materials used [399]. Pulse trains with an asymmetric temporal profile were generated by the Fourier synthesis method by Englert et al., and it was found that electron ionization in fused silica and sapphire depends on the type of the pulse trains, which are useful in tailoring laser processing for high bandgap materials at the nanometer scale [400-402]. Ahn et al. observed that the aspect ratio of femtosecond laser drilled holes in glass can be increased by using secondary pulses at 100 to 1000 ps delays [403]. High-aspect-ratio channels can also be obtained using pulses at high repetition rates, which can induce the heat accumulation effect [404]. The mechanism of the increased aspect ratio was investigated by Götte et al. using temporal Airy pulses [405]. It was shown that delayed energy deposition can achieve larger laser penetration depth compared to single pulse processing.

Various groups also use two or multiple pulses each of which has a different wavelength. Because materials often have wavelength-dependent reflection and absorption, choosing wavelengths for individual pulses in the pulse train is another dimension to tailor laser material processing. Gyamfi et al. studied the laser damage threshold of dielectric high-reflection coatings for a sequence of two pulses with different wavelengths, the fundamental 780 nm wavelength and its second harmonic at 390 nm [406]. Their work revealed the role of conduction-band electrons and their effect on laser damage threshold. Yu et al. used a pair of femtosecond laser pulses at 266 and 800 nm to investigate multiphoton and avalanche ionization [407-409]. It was shown that these two ionization pathways can be controlled individually by utilizing the wavelength-dependent absorption, and nanostructures can be fabricated more efficiently using a combination of short and long wavelengths.

5.2.2 Spatial beam shaping

It has been recognized that laser beams with a conventional Gaussian spatial profile are not suitable for some laser processing applications. Researchers have been experimenting with non-Gaussian beams and choose the optimal beam shapes suitable for a particular application. There is a large body of work on spatial laser beam shaping, and an overview of this subject can be found in [410, 411]. This section covers recent studies on applying spatial beam shaping for ultrafast laser processing of materials. The devices used for spatial beam shaping can be categorized into two types, passive devices (such as lenses and diffractive optical elements) and active devices (such as liquid-crystal spatial light modulators and deformable mirrors). Beam shaping is mostly achieved by modifying the amplitude or phase or both of the incident Gaussian optical field, either through iterative algorithms such as the Gerchberg-Saxton algorithm [412], or forward methods based on *a priori* knowledge of the relationship between the

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original and the resultant optical field. Depending on the arrangement of the optics, the final optical field can be an image (often scaled down) of a mask placed in the beam path, or the Fourier transform of the mask. The most commonly used non-Gaussian profile is the so-called "top hat" or ("flat top") beam that has a uniform intensity distribution on the focal plane. Top hat beams are preferred in laser processing because the uniform intensity will not cause the center part of the Gaussian beam to create nonuniform processing results. Sanner et al. used a nonpixelated optically addressed liquid-crystal light valve to create top hat beams (among other beam shapes), and femtosecond laser drilling using the generated beams was demonstrated as shown in Fig. 28 (a-b) [413, 414]. Squared and circular top hat beams are generated by Li et al. using a spatial light modulator in an image-based configuration [415]. In that study, ultrafast laser machining was performed with titanium samples, and the machined features were found to be consistent with the designed beam shape.

Another type of beam shape that has drawn attention lately is the Bessel and annular beam. Bessel beam belongs to the propagation-invariant optical field whose transverse intensity distribution stays the same along the axial (beam propagation) direction [416], whereas annular (or doughnut) beams can be generated using the Bessel mode or other optical fields that have an intensity null at the center. If the location of the intensity null also has a phase singularity, those annular beams are often called vortex beams or beams with orbital angular momentum. One can refer to Duocastella and Arnold for a review on using these beams for material processing [417]. Recently, Wetzel et al. reported femtosecond laser ablation of graphene using vortex Bessel beams [418]. Damage morphologies can be controlled by single and a sequence of pulses that have different topological charges (orders of the phase singularity). The same type of beams were used by Sahin et al. to study femtosecond laser processing of Au/Cr thin films deposited on quartz substrates with different laser wavelengths and Bessel beam cone angles [419]. In a study by Cheng and Polynkin, self-focusing and beam deformation were shown to be important in femtosecond back-surface ablation of borosilicate glass [420]. The propagation of high-intensity Bessel beams in dielectrics and the associated nonlinear effects were studied by Xie et al. with the aim to control internal structural modification [421]. Using the zero-order Bessel beam, cutting of soda-lime glass and sapphire was demonstrated by Lopez et al. as shown in Fig. 28 (c-d) [422]. The side walls of the cut surface have good smoothness, and the side lobes of the Bessel beam have to be taken into consideration even though they have lower intensities than the central lobe. While Bessel beam cutting of glass has been successively demonstrated, applying femtosecond Bessel beam to cut semiconductor materials is still challenging. Grojo et al. used an optical parametric amplifier (OPA) to generate femtosecond laser pulses at a wavelength of 1300 nm, which is transparent for silicon [255]. Their results show that the intensity reached inside silicon is below the damage threshold due to strong nonlinear absorption, which also deformed the Bessel beam profile at high intensities. Superposition of Bessel beams with higher orders can create petal-like beam patterns that still possess the nondiffracting property [423]. These beam shapes have been used in femtosecond laser scribing of glass and metal thin films [424] and two-photon polymerization [425]. A recent review by Stoian et al. gives an account of recent advances in femtosecond Bessel beam processing of materials [426].

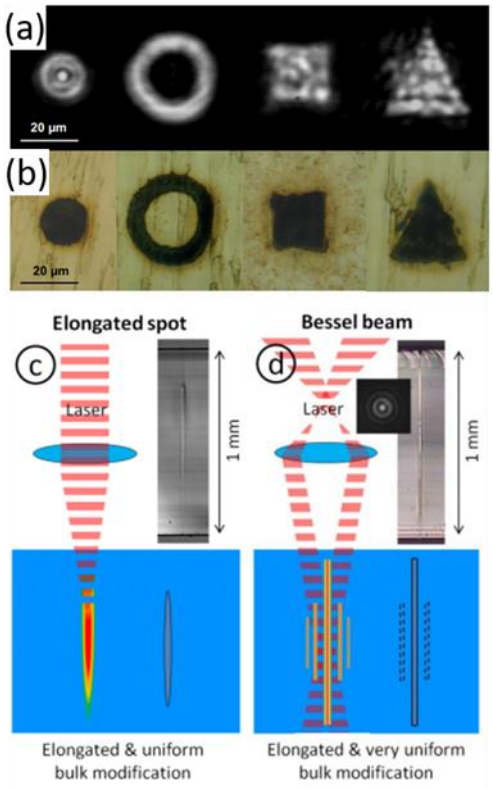

Fig. 28 Examples of material processing using spatial beam shaping (a-b) on the transverse plane to create patterns (disk, annular, square, triangular) [414], and (c-d) in the axial direction to create Bessel-like beams [422].

5.3 Laser material processing with new light sources

Lasers used in ultrafast laser material processing typically are solid-state or fiber lasers that output ultrashort pulses with pulse duration of 100 fs to 10 ps, and fundamental wavelength in near infrared (800 to 1000 nm). With the development of new laser sources, researchers have been exploring new laser-matter interaction regimes which may lead to unprecedented processing results.

5.3.1 Laser induced damage by few-cycle laser pulses

Laser induced damage by few-cycle laser pulses (one optical cycle at 800 nm wavelength is 2.67 fs) was experimentally studied in the 1990s [427, 428]. The mechanism of damage formation, such as multiphoton, tunneling and avalanche ionization, was investigated, and the application for precision laser machining was predicted. Morphological changes of silicon including amorphization, melting, re-crystallization, nucleated vaporization, and ablation were studied with pulse duration down to 5 fs [429]. With the increasing availability of few-cycle pulsed lasers, more and more research has been conducted in this pulse duration range

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towards a better understanding of laser induced damage. Sanner et al. created 7 fs pulses by self-phase modulation in a hollow-core fiber followed by pulse compression using chirped mirrors, and found a correlation between the Keldysh parameter [430] and the determinism of the damage threshold, suggesting that tunneling ionization is predominant for few-cycle pulse durations [431]. Du et al. used few-cycle, UV pulses to study free carrier dynamics in dielectric coatings, and found that free carriers in the conduction band will relax into self-trapped excitons, which will cause the incubation effect under repetitive radiation [432]. Utéza et al. studied laser ablation of fused silica in terms of ablation depth and diameter over pulse durations from 7 to 450 fs, and concluded that short pulses can achieve high precision in axial material removal [13]. In a recent study, few-cycle pulses have been used to study laser-induced periodic structures [433]. It is found that shortening the pulse duration to a few optic cycles produces surface morphology very different from many-cycle pulses. The mechanism of few-cycle laser pulse induced damage was studied by Kafka et al. in vacuum and air with single and multiple pulses [434], and a difference between the ambient environments was observed for the morphology of multiple pulse induced damage. It has been pointed out that care must be taken when handling few-cycle laser pulses due to their high peak power and broad spectrum [435, 436]. In particular, the assumption of quasi-monochromatic light, which is often used in describing electromagnetic waves, needs to be revisited for few-cycle pulses.

5.3.2 Laser induced damage at new spectral range

The spectral range that can be used for ultrafast laser material processing has been extended from the typical UV-visible-near infrared to extreme ultraviolet (XUV) and mid infrared (MIR). XUV pulses can be generated by high harmonic generation (HHG), and MIR pulses are typically generated by optical parametric amplification (OPA). Poly(methyl methacrylate)(PMMA) was ablated using XUV radiation with pulse duration of 30-100 fs from a free-electron laser [437]. The ablated surfaces exhibited good surface quality, and the results were compared with nanosecond and picosecond XUV ablation. Using an XUV/visible-NIR dual-beam setup, Mocek et al. and Jakubczak et al. investigated the surface structuring of amorphous carbon and PMMA [15, 438]. The longer wavelengths (visible - NIR) can interact with the XUV pulse induced carriers, so the absorption is enhanced. Femtosecond laser induced damage in germanium (Ge) was studied by Malik et al. using an MIR wavelength of 3.9 μm [14]. Because Ge has low linear absorption at this wavelength, this study reveals nonlinear effects that will be of interest to laser processing applications. Recently, Austin et al. conducted single-shot laser damage experiment on Ge using a wavelength range from 0.78 to 3.6 μm [439]. It was found that the oxide layer together with a thin layer of Ge are removed by the laser beam, and a new model was proposed to address the discrepancies between the currently used models.

5.4 Scalable nano-manufacturing by ultrafast lasers

Ultrafast lasers have shown superior capabilities in nano-scale manufacturing with high resolution, high quality, simple setup, and low request to the environment. To speed up the industrial application and commercialization of these techniques, it is essential to improve their scalability and fabricate nanostructures at an industrially relevant scale. The advance of the techniques of high speed motion stage and high speed galvanometer/ polygonal scan head have significantly increased the processing speed for cutting, scribing, etc. However, more technical barriers need to be overcome to make these techniques scalable.

One approach to improve the scalability and throughput is to process multiple features simultaneously. Multiple parallel pulses can be generated by different techniques, such as diffractive optical elements [440], microlens array [441], periodical binary phase mask [442], and spatial light modulator [149, 443, 444]. The multiple parallel beams have been used to generate complex 2D and 3D structures on different materials. This method can efficiently reduce the processing time for high volume processing of the same feature.

Another effective method is to use non-diffractive Bessel beams instead of the commonly used Gaussian beams, as described in Section 5.2.2. Ideal zero order Bessel beams are formed by axicon lenses from the axially-symmetric interference of two plane waves to yield a high intensity central core surrounded by lower intensity concentric rings. With the same beam diameter, the Bessel beam can maintain high intensity over a distance much longer than the Rayleigh length of a Gaussian beam. Therefore, the Bessel beam is very efficient to fabricate high aspect ratio nanochannels [445-447]. Fig. 29 shows a channel with a diameter of 400 nm and a length of 43 μm created by a single Bessel pulse in glass. It should be pointed out that the Bessel beam is only effective to treat transparent materials to take advantage of its long intensity distribution length.

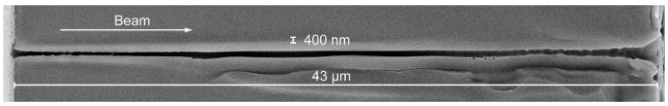

Fig. 29 A high aspect ratio nanochannel fabricated by a single femtosecond Bessel pulse [447].

As described in Section 4.2, laser surface micro/nanostructuring could be a highly scalable technique. The nanoscale laser-induced periodic surface structures are formed and self-organized within the whole laser irradiation area. They are created within a few hundreds of ps, and do not need super focusing of the laser beam. With the help of high speed laser scanning head, nano-scale surface structures can be potentially fabricated on a 1 m^2 surface within 30 mins by a high repetition rate ultrafast laser (assuming 1.5 m/s scanning speed, 2 kHz repetition rate, 1 mm spot diameter, and 50% overlapping ratio).

5.5 Other emerging application areas of ultrafast laser based manufacturing

Ultrafast laser-based manufacturing is becoming a powerful and comprehensive platform for various applications. There are many innovative techniques invented for manufacturing emergent micro-to-nano devices and addressing

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the technical challenges of modern life. These fields include, but are not limited to, the following.

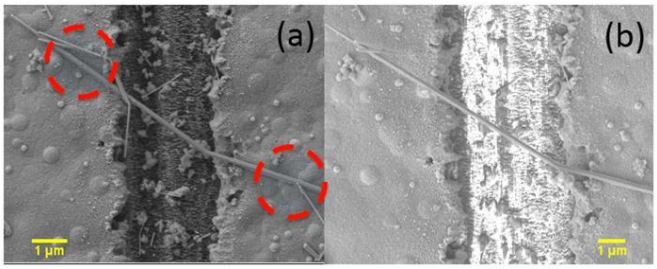

Fig. 30 Femtosecond laser joining of single Cu nanowire. The silver film width is 4 μm. (a) Before joining. Two dashed circles indicate the joining site. (b) After laser joining. The thin Cu nanowires were blown away by laser illumination.

Ultrafast laser-based nanojoining is emergent as a crucial manufacturing method for nanodevices [448, 449]. Nanojoining is an essential step to construct nanodevices through joining nanoscopic building blocks with strong bonds (metallic or covalent bonds, not a weak bonding force like Van der Waals, hydrogen bonds, or capillary force) and forming permanent bonding. It is important to point out that nanojoining is different from selected growth, which is a popular method to assemble nanomaterials, such as homogenous and heterogeneous nanowire junctions [450]. Nanojoining allows bottom-up manufacturing and assembles hybrid structures by joining dissimilar nanomaterials, maximizing the unique properties of nanomaterials. Femtosecond laser-induced sintering of silver nanoparticles has been demonstrated [451]. The experiment displayed that a very low laser fluence down to 900 μJ/cm^2 is needed to melt silver nanoparticles. The self-localized joining mechanism is similar to plasmonic joining, in which silver nanowires are melted and joined at hot-spots due to localized plasmonic excitation [452, 453]. It has been widely shown that nanoparticles can melt at a lower temperature than the bulk melting point due to size effects; molecular dynamics simulation predicts surface melting that also occurs at a lower temperature than the melting point of nanoparticles [454, 455]. This result indicates that the sintering of nanomaterials can be induced by surface melting. Recently, silver nanowires were also joined with a focused 532 nm CW laser [456, 457] and femtosecond laser [458]. Controllable multilevel memory is demonstrated in joined titania nanowires [459]. Moreover, a super-sensitive glucose sensor was fabricated by joining a single copper nanowire to silver film [460]. Shown in Fig. 30, a copper nanowire bridges a silver film gap cut by laser beam. After joining the two ends of the copper nanowire, electronic resistance decreased from 1 kilo-ohm to 200 Ohm. Such a joined single copper nanowire can sense glucose at a concentration as low as 10^{-6} M [460]. It is thus reasonable to predict that more applications of joined nanodevices can come to real world applications with further maturation of laser-based nanojoining.

New materials and new phases synthesized via femtosecond laser-induced micro-explosions have also appeared as a hot research topic [461]. Electron ejection due to ultrafast laser-matter interaction will lead to a Coulomb explosion of positively charged ions after laser-induced vaporization of electrons. This Coulomb explosion is confined in a microscale space in the vicinity of the focal spot. High-temperature and high-pressure conditions can be used for the synthesis of new materials, as well. Hu reported that *sp*-bonded carbon chains can be formed on a graphite surface using femtosecond laser irradiation associated with the formation of amorphous tetrahedral carbon (diamond-like carbon) [462]. This *sp*-bonded carbon chain species can be precursors for various carbon allotropes, including carbon nanotubes, fullerenes and carbynes [463]. Rapp reported a new tetragonal polymorph of silicon induced by confined microexplosions [464]. On the other hand, laser peening induced by fs laser pulses can be used as a powerful surface treatment tool for metals [465]. Femtosecond laser peening research was motivated by the process simplicity and high shock pressure attainable with fs pulses in comparison to ns laser peening. More recently, encouraging results in terms of compressive residual stress and hardness enhancement were reported in fs laser peening of aluminum [466] and steel [467] free from a protective coating and a transparent overlay. Hence, it can be said that ultrafast laser peening has shown promising results but more research is needed in this area.

6. SUMMARY AND FUTURE RESEARCH NEEDS

This comprehensive review exposes a wide range of research activities utilizing ultrafast lasers, with a focus on fs laser applications in manufacturing processes. From surface structuring to machining in bulk to scalable manufacturing, fs lasers have been shown to be a crucial tool in the world of precision micro and nano machining for a wide variety of materials, ranging from metals to dielectrics to biomaterials. Since the inception of high power ultrafast lasers over three decades ago, various advanced ultrafast light sources have emerged today and become more powerful (e.g. TW level), more colorful (from XUV to mid-IR), much shorter in pulse duration (e.g. attoseconds), and more economical (e.g. fs fiber laser), which opens up many new opportunities in manufacturing process innovation. Further down the road, here are some future research needs in ultrafast laser based manufacturing.

(1) Laser-matter interaction under irradiation with ultrahigh intensities, few cycle pulses, mid-IR wavelengths
(2) Micro/nano machining using shaped beams
(3) Laser induced material phase synthesis
(4) Additive manufacturing at micro/nano scale (e.g. nanojoining)
(5) Micro/nano machining at industrially relevant scale
(6) Micro/nano surgery
(7) Nanowelding and nanojoining

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